PAPER

Setup of high resolution thermal expansion measurements in closed cycle cryostats using capacitive dilatometers

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Abstract
We present high resolution thermal expansion measurement data obtained with high relative sensitivity of $ΔL/L = 10^{-9}$ and accuracy of ± 2 % using closed cycle refrigerators employing two different dilatometers. Experimental details of the set-up utilizing the multi-function probe integrated with the cold head of two kinds of closed cycle refrigerators, namely, pulse tube and Gifford-McMahon cryocoolers, has been described in detail. The design consists of decoupling the bottom sample puck and taking connections from the top of the multi-function probe to mitigate the vibrational noise arising from the cold heads, using which smooth and high quality thermal expansion data could be obtained. It was found that dilatometer #2 performs a better noise mitigation than dilatometer #1 due to the constrained movement of the spring in dilatometer #2. This was confirmed by finite element method simulations that were performed for understanding the spring movement in each dilatometer using which the effect of different forces/pressures and vibrations on the displacement of the spring was studied. Linear thermal expansion coefficient $α$ obtained using both dilatometers was evaluated using derivative of a polynomial fit. The resultant $α$ obtained using dilatometer #2 and either of the closed cycle cryostats on standard metals silver and aluminium showed excellent match with published values obtained using wet cryostats. Finally, thermal expansion measurements is reported on single crystals of two high temperature superconductors YBa$2$Cu$_3$−$x$Al$_x$O$_6+$δ and Bi$_2$Sr$_2$CaCu$_3$O$_{8+x}$ along the c-axis with very good match found with published data obtained earlier using wet liquid helium based cryostats.

1. Introduction
Thermal expansion is an important thermodynamic technique to probe phase transitions in solids [1–4]. Volume thermal expansion coefficient, $β$, describes the change in volume of a material in response to a change in temperature while the linear thermal expansion coefficient, $α$, describes the corresponding change in the length of a given material. $β$ is defined as:

$$β = \frac{1}{V} \left( \frac{∂V}{∂T} \right)_p$$

and

$$\frac{1}{L_x L_y L_z} \left( \frac{∂(L_x L_y L_z)}{∂T} \right)_p$$
\[ \alpha_x + \alpha_y + \alpha_z \]

For an isotropic material,

\[ \alpha_x = \alpha_y = \alpha_z = \alpha \]

\( \Rightarrow \beta = 3\alpha \)

Grüneisen parameter, defined as \( \Gamma = -\frac{\partial \ln V}{\partial \ln \rho} \), where \( \rho \) is the isothermal compressibility, relates the volume thermal expansion coefficient \( \beta \) to specific heat \( C_V \) [5, 6]. Zhu et al defined a generalised Grüneisen parameter \( \Gamma - \frac{\beta}{\alpha} = -\frac{1}{V^C} \int_0^T \frac{\partial f}{\partial V} ds/\partial T \) for probing quantum phase transitions where the Grüneisen parameter is expected to diverge [7]. Such divergences in thermal expansion have, in fact, been observed in quantum critical materials like \( Sr_3Ru_2O_7 \) [8], \( Ce(Fe_2N)_3CuBr_4 \) [9], \( CeCoIn_5 \) [10] etc. Similarly, thermal expansion has been used as an important technique for studying correlations with onset of superconductivity to structural transitions in the crystalline lattice [11–17]. In this regard, thermal expansion measurements in high temperature superconductors (HTSC’s) are very valuable since the transition from the normal Mott insulating state to the superconducting state is known to be accompanied by a corresponding structural transition from the tetragonal state of Mott insulator to orthorhombic state of superconductors [18, 19]. Additionally, thermal expansion measurements in the HTSC \( YBa_2Cu_3O_{6+\delta} \) (YBCO) have shown that superconductivity in YBCO is not only effected by the oxygen content of the superconductor but also by the degree of oxygen order in the CuO chain layer [14]. Since HTSC’s are proposed to be governed by an underlying quantum critical point [19–22], thermal expansion measurements - a generalised Grüneisen parameter, offer a very important tool to probe quantum criticality in HTSC’s. Of the many available techniques to measure thermal expansion of solids, capacitive dilatometry is known for its ability to resolve relative length changes of \( \Delta L/L < 10^{-9} \), such that a resolution of \( \Delta L = 10^{-2} \AA \) can be achieved in samples of lengths in the mm range [2, 3, 23–34]. In comparison, a resolution of \( \Delta L = 10^2 \AA \) is reached using optical methods [35] and that of \( \Delta L = 10^{-1} \AA \) is reached using the piezocantilever technique [36]. In the capacitive dilatometry technique, the sample to be measured and two parallel metal plates are in a configuration such that a length change \( \Delta L \) of the sample results in a change in the gap between two metal plates placed parallel to each other. A high resolution capacitance bridge is, then, used to measure the capacitance between the plates before and after the length change, represented as \( C_i \) (initial capacitance) and \( C_f \) (final capacitance) respectively. The change in length \( \Delta L \) is equal to the negative of the change in gap between the capacitor plates, \( \Delta D = \epsilon_0 \epsilon_r A \left( 1/C_i - 1/C_f \right) \), where \( \epsilon_0 \) and \( \epsilon_r \) are absolute and relative permittivity of the medium in between the plates and \( A \) is the area of the capacitor plate. For a capacitive dilatometer to be useful for studying thermal expansion at low temperatures and high magnetic fields it must have certain features: (1) low thermal mass, (2) relative insensitivity to magnetic fields, and most importantly (3) good resolution. Low thermal mass ensures that the dilatometer can be efficiently cooled, requiring minimum amounts of helium in a cryogen based system or energy spent by a cryocooler in a dry system. This can be achieved by reducing the dimensions of the capacitor plates and the flanges that hold the capacitor plates, sample platform etc. Superconducting samples or the systems exhibiting quantum phase transitions are frequently millimeter sized single crystals. Their interesting thermal expansion characteristics are observed at low temperatures where the thermal expansion is quite small. So, one requires dilatometers that have high resolution, typically in the sub-angstrom resolution range, to ensure that the small changes in thermal expansion can be picked up by the dilatometer efficiently. The first description of such a miniaturized dilatometer was given by White [23], which was later followed by Pott and Schefzyk [24], Swenson [25] and many others [12, 26–34]. The designs can be differentiated based on three important characteristics: (1) material used in different components of a given dilatometer (2) sample mounting architecture, and (3) capacitor plate movement. A dilatometer that can be used to perform measurements in high magnetic fields requires its parts to be manufactured with materials that are insensitive to magnetic fields. For its relative insensitivity to magnetic field, ease of machinability and well known thermal expansion characteristics, Oxygen free high conductivity (OFHC) copper has been used in Schmiedeshoff’s dilatometer design [31], while Küchler et al [34] used an alloy of beryllium and copper to construct their dilatometer. On the other hand, Neumeier et al [32] constructed their dilatometer using quartz to minimise the contribution due to background as much as possible. The initial dilatometer designs were such that the samples were placed between the plates of the capacitor putting severe restrictions on the sample shape or geometry [23, 24]. However, designs of Schmiedeshoff et al [31] and Küchler et al [34] have an open architecture, wherein, the sample is placed outside the capacitor plates resulting in choice of sample shape size and mountings. Finally, parallel plate movement of the capacitor plates have been achieved both by Neumeier et al [32] and Küchler et al [34] by using leaf springs. Thermal expansion measurements using capacitative dilatometry typically use wet cryostats where liquid helium is used as a cryogen [37, 38]. The reason for this usage is the sub-angstrom level resolution that one needs to achieve in order to investigate low temperature phase transitions in small sized single crystals. Closed cycle refrigerators (CCR’s), on the other hand, frequently suffer from noise imparted to the experimental probe via
the mechanical vibrations arising from the moving part of the CCR [39–42], thereby, seriously limiting the usage of CCR’s for thermal expansion measurements. However, with the rapid improvement in the CCR technology in the last 20 years, lower noise vibration levels have been achieved in the CCR’s [41, 42], giving hope for their usage in noise-sensitive measurements like thermal expansion and reducing the dependence on the ever-depleting and expensive cryogen of liquid helium. In this work, we present thermal expansion measurements on CCR’s and show excellent data quality that matches with the data obtained via wet cryostats, demonstrating technological advancement for future thermal expansion measurements that can be done using CCR’s wherein similar quality data as that obtained using conventional liquid helium based wet cryostats could be obtained using CCRs. The CCR’s chosen for the measurement were of two kinds: (i) Gifford-McMahon (GM) CCR and (ii) Pulse-Tube- (PT) CCR. These CCR’s were implemented in the widely used physical property measurement system (PPMS) from Quantum Design Inc. Thermal expansion measurements on these CCR’s were done on two different dilatometers, one based on Schmiedeshoff’s design [31] that was fabricated, assembled and set-up in house, and the other based on Küchler’s design [34], obtained commercially. The performance of both the dilatometers in the different CCR’s was checked with standard metal samples of copper, aluminium and silver, with well-known thermal expansion characteristics. We find that thermal expansion measurement data obtained using either of the CCR’s and either of the dilatometers give excellent quality data, comparable to those obtained using wet liquid helium cryostats. However, comparison between the cryostats and dilatometers reveal that PT cooled CCR and Küchler’s dilatometer give better data. In order to investigate the reason for the difference of data quality between Schmiedeshoff’s dilatometer and Küchler’s dilatometer, we performed finite element method (FEM) simulations on the spring movement in both the dilatometers under the action of a force. The simulations confirmed that a better noise mitigation happens in Küchler’s dilatometer as compared to Schmiedeshoff’s dilatometer. Finally, thermal expansion measurements of two different superconducting crystals YBa_{2}Cu_{3−x}Al_{x}O_{6+δ} (Al-YBCO) and Bi_{2}Sr_{2}CaCu_{2}O_{6+δ} (BSCCO-2212) are reported along the crystallographic c-axis which revealed very good match with previously published data obtained using wet cryostats.

The paper starts with the description of the experimental set-up where the PPMS architecture comprising the multifunction probe and the bottom sample puck of the PPMS is described. This is followed by a description of Schmiedeshoff’s dilatometer and Küchler’s dilatometer. A detailed description is, then, made of Gifford-McMahon CCR and the integration of the cold head of the CCR with the PPMS cryostat. We show the bad quality data that arises when the bottom sample puck is used along with multifunction probe of PPMS. The next part of the experimental set-up describes the noise mitigation that is achieved by decoupling the bottom sample puck and taking connection from the top of the multifunction probe. In the next section of the paper, we present a detailed description of calibration of Schmiedeshoff’s dilatometer and calculation of the empty cell effect for both the dilatometers which is very important to estimate the background signal arising from the dilatometers. This is followed by measuring thermal expansion of two standard metals, namely, copper and silver. Here, we describe two different methods of calculation of \( \alpha \) from the relative length change, the first being a simple numerical derivative, and the second a polynomial fit. Both the methods yield different levels of noise in the calculated \( \alpha \) and we find it necessary to consider both the methods of calculating \( \alpha \) to better understand the CCR’s contribution to the thermal expansion. The next section describes the details of finite element simulations that were done on the springs of both the dilatometers. Here, effect of variation of radial distance on the resultant displacement of the spring was studied as a function of thickness of the spring as well as applied force. The final section describes thermal expansion data obtained on the single crystals of two high temperature superconductors YBa_{2}Cu_{3−x}Al_{x}O_{6+δ} (Al-YBCO) and Bi_{2}Sr_{2}CaCu_{2}O_{6+δ} (BSCCO-2212).

2. Experimental setup

The workhorse of Quantum Design Inc’s PPMS is what is called as the sample ‘puck’ which is a unique modular component that provides great flexibility in performing different types of measurements on a PPMS like electrical resistance, thermal conductivity, vibrating sample magnetometry etc [43, 44]. Figure 1(a) and (b) show the top and bottom view respectively of the sample puck. The top view shows a blank puck where no electrical contact pads are present. The bottom view shows a socket assembly terminating in 12 pin female connector leads that gets connected to the male pins of the sample chamber which is pre wired to the system electronics. Some of the 12 pins have connections to the sample chamber heater while others are free to be used for user specific experiments. A thick braided wire containing all the connections to the 12 pin connector can be seen arising from the bottom portion of the puck in figure 1(b) (red coloured wire). The actual wire can be seen marked in figure 1(d). So, when installed in the PPMS sample chamber, the ‘puck’ provides the sample with both electrical and thermal contact to the sample chamber via the braided wire.
The multifunction probe (MFP) of Quantum Design Inc. is a versatile insert that comes with radiation shields, wires, and a space where user-specific instruments can be integrated. Figure 1(c) shows the bottom-most part of the MFP that comprises a blank sample puck at the bottom where user-specific electrical contacts can be made. In the first attempt to use the MFP for doing thermal expansion measurements, we connected our dilatometer (marked in figure 1(c)) to the frame of the MFP via a copper adaptor and screws. The electrical leads from the dilatometer (blue wire in figure 1(c)) were, then, soldered onto a resistance puck (shown as green rectangle with contact pads in figure 1(c)) of the PPMS which was soldered to the sample puck via the thick
braid wires, shown clearly on an expanded scale in figure 1(e). Figure 1(d) shows an actual image of the final assembly. In order to get a capacitance data, we connected the resistance bridge outside the cryostat to the capacitance bridge. When inserted inside the cryostat, the dilatometer assembly inside the MFP can give capacitance data as a function of temperature.

In this paper, we describe the details of two different dilatometers that we used for thermal expansion measurements. The first one, called as dilatometer #1, was designed based on Schmiedeshoff’s work [31]. Figures 2(a) and (b) show a computer realisation of dilatometer #1 and dilatometer #2 respectively made using CREO Parametric software. Dilatometer #1 consists of an upper capacitor plate e that is fixed and a moving lower capacitor plate g. The two capacitor plates are separated by a shim f. As can be seen from figures 2(a) and (b), both dilatometer #1 as well as dilatometer #2 have an open architecture design of sample mounting where the sample S is placed outside the capacitor plates. Length changes brought in the sample due to a temperature variation is manifested as a change in the distance between the two capacitor plates brought about efficiently through a spring (k in dilatometer #1 and K in dilatometer #2). All the parts were fabricated using OFHC Copper and assembled together according to the details given in reference [31]. After fabricating the components, they were gold plated to protect them from oxidation. Figure 2(c) shows an actual image of the assembled dilatometer. Dilatometer #2, on the other hand, was obtained commercially from Kuechler Innovative.

Figure 2. Schematic of (a) dilatometer #1 based on [31]. a represents main flange, c is the lock ring, d sample platform, S sample, e upper capacitor plate, g lower capacitor plate, f shim, m screws, j mounting plate and k spring (b) Schematic of dilatometer #2 based on [34]. S is the sample, A is the main flange, B the guard ring, L top capacitor plate, H bottom capacitor plate, K leaf spring, P adjustment screw, N nut and Q adapter. (c) is the assembled dilatometer corresponding to the schematic (a) while (d) is the image of the actual dilatometer corresponding to the schematic (b).
Measurement Technology, Germany, which is based on the design of reference [34] and is shown in figure 2(b). It has a fixed lower capacitor plate $H$ that is mounted onto the cell frame $A$, a movable upper capacitor plate $L$ and an adjustment screw $P$ using which the sample $S$ is fixed. The movement of the upper capacitor plate is brought about by a set of leaf springs $K$ machined out of BeCu. From the figure 2(b), it is clear that the cell frame, the two capacitor plates as well as the spring are machined out of a single block of BeCu, resulting in a very high level of parallelism between the two capacitor plates. Figure 2(c) shows an actual image of the dilatometer that was mounted onto the MFP of the PPMS.

It is a very well known fact that liquid helium is a very fast depleting resource of the world. Its shortage and, consequently, high prices has been plaguing both experimental physicists as well as chemists who rely on liquid helium for performing low temperature experiments, leading to shut-downs of their equipments due to lack of liquid helium [37]. In fact, according to an estimate, the world would run out of liquid helium by 2030 [38]. So, closed cycle refrigerators (CCR’s), where the amount of refrigerant is approximately a constant, is the need of the hour. In fact, CCR’s are now very commonly used for performing experiments like resistivity, heat capacity, thermal conductivity etc. However, noise sensitive experiments like thermal expansion, scanning tunneling microscopy etc still do not use CCR’s since the vibrations arising out of CCR’s are detrimental to the data. In this work, we have incorporated the two dilatometers described above in two different kinds of CCR’s to be able to achieve sub-angstrom resolution thermal expansion data. To this end, we used two different PPMs’s that employ two different CCR’s for providing the cryogenic environment: (1) PPMS-Evercool and (2)PPMS-Dynacool [43, 44]. PPMS-Evercool uses Gifford-McMahon (GM) CCR which is named as CCR#1. On the other hand, PPMS-Dynacool employs a pulse-tube (PT) CCR which is named as CCR#2.

Figure 3(a) shows a schematic of a two stage GM cryocooler, CCR#1, that was used in our experiments. A CCR, typically, has a cold head or an expander and a compressor. Cold head is the place where refrigeration takes place and is connected to the compressor via high and low pressure gas lines [39–41]. The compressor (not shown in figure 3) has a piston compressor whose motion produces the required pressure pulses, thereby, providing the necessary helium gas flow rate to the pressure lines, and was kept at more than 20 feet away from the cold head. The cold head comprises regenerators, heat exchangers, expansion volumes and displacers. Pressure pulses results in volumetric change of the working helium gas in the expansion volume of the cold head. The functioning of regenerators, heat exchangers, expansion volumes and displacers together results in achieving low temperatures. The source of mechanical noise to the system is through the movement of the displacer that shuttles the helium gas back and forth from the cold end to the warm end in CCR#1. Using a second heat exchanger, regenerator and displacer (two-stage cryocooling), a base temperature of 4.2 K is achieved in CCR#1. A PT based cryocooler, CCR#2 (not shown), has an advantage over CCR#1 as far as moving parts are concerned since CCR#2 doesn’t have moving parts at the cold end. However, a gas pulse still flows in it, thereby, injecting mechanical noise in the system [39–41].

![Figure 3](image-url)
Figure 3(b) shows the integration of the cold head of the CCR#1 with the PPMS cryostat. $K_1$ and $K_2$ represent the two stages of the CCR#1 described in figure 3(a). When the working gas, namely, helium, is constrained to move through the first stage and the second stage, it cools at all the stages and is finally condensed at the second stage. The condensed liquid is then collected at the bottom of unit and is made to reach the sample stage via a pipe (dark blue coloured in figure 3(b)). Figure 3(b) also shows the MFP that has the dilatometer connected to the sample puck via the resistance puck (refer to discussions related to figure 1). When electrical connections were taken using this assembly, the resultant capacitance versus temperature graph, as shown in figure 3(c) consisted of only noise in the form of capacitance spikes. At each such instance of capacitance spike, the capacitance bridge (described below) showed a large loss value.

We measured capacitance as a function of temperature many times and always found similar spikes in the capacitance data. However, for all other measurements possible in a PPMS, namely resistivity, specific heat etc where the connection to the PPMS is via the bottom sample puck, we never encountered any noise in the data similar to those shown in figure 3(c). This confirmed that the source of noisy spikes in the capacitance data was the vibrations arising from the cold head of CCR that get coupled to the dilatometer via the bottom sample puck. Barton et al found that vibrational noise arising from CCR’s affect low capacitances (of the order of few pF) of their system [45]. Since the starting capacitance of our dilatometers are always in the range of 12-15 pF, it was clear that this technique of measuring thermal expansion would always give us noisy data. In an effort to make highly sensitive gravitational wave interferometers, Caparrelli et al were able to reduce the vibrations arising out of their PT cryocooler by mechanically decoupling the cryostat flange from the cryocooler cold head using special silicon bellow[46]. Guided by this, we decided to try decoupling the cryostat from the cold head of CCR by removing the bottom sample puck altogether and take connections directly from the top of the MFP.

Figure 4(a) shows the bottom-most part of the MFP where the bottom sample puck was removed. Since the bottom sample puck is removed, the present design avoids the PPMS sample chamber wiring. So, in order to get electrical connections to the dilatometer, we routed the sample wiring through a pair of ultra-thin coaxial cables, marked as thin silver coloured wires in figure 4(a), through the top. In order to realise this, we drilled two holes at the top of the MFP and fitted two Lemo connectors (marked in figure 4(b)) to whose inner ends, the coaxial wires were soldered. In order to reduce the heat leak caused by the coaxial wires, they were wrapped around the entire...
length of the MFP and were heat sunk at few places via the usage of thermal anchors marked in figure 4(c). In order to shield the ultra-thin coaxial cables from the measuring capacitance bridge till the capacitor plates of the dilatometer, we used feedthroughs as well as centre-ring at the KF flange made of plastic as shown in figure 4(b). The entire MFP with the new design is shown in figure 4(c).

As described above, if the sample puck was used at the bottom, then the MFP would have been thermally coupled to the annular region at the bottom of the sample chamber via the 12 pin connector, where heaters warm the helium gas to the required temperature (refer to figure 3(b)). However, since the bottom sample puck was removed in the present design to minimise the noise vibrations, we connected our own Cernox resistance thermometer in order to get temperature measurements. The Cernox thermometer was obtained from Lakeshore Inc. commercially and was placed close to the sample position on the dilatometer frame (see figure 4(a). The cabling to MFP top was done via a twisted pair of Beryllium Copper wire and connections taken via a Fischer connector, marked in figure 4(b). The wires were wound across the entire length of the MFP and heat sunk at the thermal anchors.

The final measurement setup that was used for thermal expansion measurements is shown in figure 5. The setup consists of four different components: (1) dilatometer (2) cryostat (3) capacitance bridge and (4) temperature controller. Figure 5 (a) shows a schematic of the new design of the MFP as described in figure 4 above, integrated in the two-stage CCR#1, whose details have already been described above. The difference in this design vis a vis that of figure 3(b) is the design of the MFP, the presence of the plastic centre-ring O instead of the stainless steel centre-ring used in figure 3(b) and a set of electrical feedthrough’s F for taking electrical connections from the top. Since the bottom sample puck has been removed, our expectation is that the noise in the capacitance data would be removed resulting in a smooth continuous curve.

In order to get electrical connections to the capacitor plates in the dilatometer#1, we first soldered two magnanin wires to the top (fixed) and bottom (movable) capacitor plates [31] and then soldered the two ultra-thin co-axial wires to the magnanin wires. The coaxial wires, were then, taken to the top of the MFP as described above, and soldered to the vacuum side of the two hermetic co-axial Lemo connectors. Dilatometer#2, on the other hand, already came with a set of ultra-thin co-axial wires that were directly connected to the Lemo connectors. The two capacitor plates and a ground wire were then, connected to an Andeen-Hagerling capacitance bridge (Model AH2550A) for capacitance measurements in a three terminal capacitance fashion (figure 5(b)) to reduce stray capacitances of the cables, surroundings etc [24]. In this scheme of measurement, a fixed frequency of 1kHz from the generator G excites the ratio transformer comprising legs L1 and L2 each having taps to select precisely defined voltages that drive legs L3 and L4 of the bridge. L3 comprises known
variable capacitance $C_0$ and resistance $R_0$ generated from the bridge and is balanced to the unknown capacitance $C$ and resistance $R$ independently arising from the sample and comprise leg $L_4$ [47].

Two twisted pairs of Beryllium Copper wires carry the connections from a cernox temperature sensor that was attached to the dilatometer body (refer figure 4(a)), to a Lakeshore temperature controller (Model LS-350) as shown schematically in figure 5(c). A LabVIEW program [48] was written to control the temperature read out from the temperature controller, its scan rate as well as capacitance reading from the capacitance bridge. The automation program also plots the capacitance data continuously as a function of temperature in both CCR#1 and CCR#2. Figure 5(d) shows an external image of the PPMS cryostat showing the CCR#1 which is marked as $C$, with the helium gas cylinder marked as $H$. The top end of the MFP with its Lemo connectors are also seen. Data carrying cable from the Lemo connectors, to whose inner ends the coaxial wires from the capacitor plates of the dilatometer are soldered, to the capacitance bridge is marked as $E$ (only one of the two cables is shown). Figure 5(e) shows a representative plot of the temperature variation of the capacitance when the MFP was used according to the new design of figure 4. A fantastic improvement in the data quality is immediately apparent when compared to that obtained in figure 3(c). It can be seen that the capacitance data is now smooth and continuous without any intermittent spikes, similar to that obtained in wet cryostats.

Motivated by the smooth data quality achieved in this new design, we tested various samples and measured capacitance at different points in time to confirm data repeatability. The entire exercise of trying out the old design, coming up with a new design and testing it took us 4 years. Once we were certain that the new design works and is robust, we then, went on to do thermal expansion measurements on different samples. All samples were cleaned with ethanol and iso-propanol and sanded lightly with sand paper of various grit sizes to make two parallel and smooth surfaces before mounting in a given dilatometer for measurement, except for Al-YBCO crystal which naturally has a cube shape. Hence, for this crystal no sanding was done and it was loaded after cleaning with ethanol. The temperature was swept in a continuous mode at a rate of 0.2 K/min. The measurement time for each capacitance data point was 8 seconds in averaging mode [47]. We used a combination of two software packages, (1) OriginLAB [49] and (2) Octave/MATLAB [50] for data analysis, curve fitting and data plotting.

3. Measurement and analysis

We now describe the details of the thermal expansion measurements that were done on standard metals as well as two high temperature superconductors using the two dilatometers dilatometer#1 and dilatometer#2 installed in two different CCR’s, namely, CCR#1 and CCR#2.

3.1. Calibration and test at ambient temperature and pressure

Before mounting the samples on the dilatometers and loading them inside the CCR’s for temperature dependent capacitance measurements, it was mandatory to do some room temperature tests to check for parallelism of the capacitor plates. The room temperature test involved measuring the capacitance, $C$, as a function of distance between the plates, $D$. For an ideal parallel placed capacitor with a surface area $A$, the capacitance is given by:

$$C = \frac{\varepsilon_0 \varepsilon_r A}{D}$$

(6)

where $\varepsilon_0 = 8.8542 \times 10^{-12} F/m$ is the permittivity of vacuum, $\varepsilon_r$ is the dielectric constant of the medium between the parallel capacitor plates. If the dilatometers do have the ideal parallel placed capacitor geometry, the experimentally obtained $C$ versus $D$ curve should follow equation (6).

3.1.1. Dilatometer #1

In order to test the validity of the equation (6) and check for possible deviations from the ideal geometry, we performed a $C$ versus $D$ test at room temperature and ambient pressures for dilatometer#1. In this dilatometer, the change in distance between the plates is achieved by first shorting the sample platform $d$ to the lower capacitor plate $g$ and then rotating the sample platform by an angle $\theta$ to achieve a linear motion of the sample platform which eventually pushes the lower capacitor plate closer to or farther from the fixed upper capacitor plate $e$ depending on the direction of rotation. The precise amount by which the distance between the capacitor plates changes was measured using the formula in equation (7), wherein the symbol $\theta$ represents the angle by which the sample platform is rotated, $\theta_{\text{max}}$ is the value of the angle $\theta$ where the two capacitor plates are about to short and $w$ is a constant which represents the pitch of the sample platform thread having a value of 882 nm per degree of rotation.

$$D = (\theta_{\text{max}} - \theta) \times w$$

(7)
Substituting equation (7) in equation (6) and assuming the dielectric constant of air \( \epsilon_r \) as 1, we get:

\[
\frac{1}{C} = \left( \frac{w}{\epsilon_0 A} \right) \theta + \left( \frac{\theta_{\text{max}} \times w}{\epsilon_0 A} \right).
\]  

(8)

The equation (8) gives \( w/\epsilon_0 A \) as the slope of the plot of \( 1/C \) versus \( \theta \) curve, which must be a straight line. We fitted this equation in a least square manner to a measured variation of \( 1/C \) versus the variable \( \theta \) shown in figure 6. It can be seen from figure 6 that a straight line fits the data (shown as red filled circles) very well in the 12 pF \( \leq C \leq 21 \) pF range. Deviation from linearity in the higher angle range may be arising due to non-perfect parallel arrangement of the capacitor plates [31]. The slope obtained from the straight line fit in the 12 pF \( \leq C \leq 21 \) pF range is 0.077 41 pF \( \theta^{-1} \). So, the area of the capacitor plate obtained from the fit is 1.3338 \( \times 10^{-4} \) m\(^2\) which is \( \sim 0.05\% \) of the actual area of the capacitor plate (1.2711 \( \times 10^{-4} \) m\(^2\) \( d = 12.725 \) mm). This difference in the actual value and the obtained value is consistent with effects like edge effects for circular capacitor plates separated by a small gap (geometry of dilatometer \#1) [51], roughness and curvature of capacitor plates etc [24].

3.1.2. Dilatometer \#2

For this dilatometer, the change in the length of the sample, \( \Delta L \), is achieved by measuring the change in the capacitance \( C \) from the starting capacitance \( C_0 \) as follows:

\[
\Delta L = \epsilon_0 \pi r^2 \frac{C - C_0}{C \cdot C_0} \left( 1 - \frac{C}{C_0} \right) \frac{C_0}{C_{\text{max}}}.
\]

(9)

where \( r \) is the radius of the capacitor plate (=7 mm) and \( C_{\text{max}} \) is the value of the capacitance at which the two capacitor plates short. Equation (9) was derived by Pott and Schefzyk [24] by incorporating a slight tilt between the capacitor plates. The change in the length, \( \Delta L \), was obtained by using a dial gauge of resolution 1/100mm while the capacitance was measured using a commercial capacitance bridge. By tightening the adjustment screw \( p_a \), a graph of \( \Delta L \) versus \( C \) was obtained for different values of shorting capacitance \( C_{\text{max}} \). This calibration chart was provided to us by the manufacturer. We have operated our dilatometer \#2 between the range of values of \( C_0 \) varying between 12 pF to 14 pF which corresponds to a length change of 8.7372 \( \mu \)m for a capacitance change of 1 pF for a sample of length 1 mm. In this range of capacitance values, a parallel plate geometry assumption holds true [34]. Dilatometer \#2 has incorporated a parallelogram suspension mechanism [27, 34] for achieving the parallelism between the capacitor plates. We have studied this as well as other aspects of such a design by performing a FEM simulation, the details of which are presented in section 3.

3.2. Empty cell effect

In a thermal expansion or magnetostriction measurement, the sample as well as the various components of the dilatometer cell undergo changes in their dimensions in response to a change in temperature or magnetic field. This means that the measured length change of a sample, \( \Delta L_{\text{sample}} \), is the difference between the change in length of the sample, \( \Delta L_{\text{sample}} \), and a background contribution arising from the change in length of the
dilatometer cell, $\Delta L_{\text{cell}}$, known as the cell effect \cite{31, 34}. The cell effect can be determined by performing a thermal expansion measurement on a reference sample, whose thermal expansion characteristics are well known. For both dilatometer #1 as well as dilatometer #2, the sample platform itself could be used as reference samples. This was possible since dilatometer #1 was fabricated from OFHC copper whose thermal expansion is well known and for dilatometer #2 which was machined from Cu$_{1-x}$Be$_x$ with a low beryllium content of 1.84, copper's thermal expansion data could be used \cite{34}. For the measurements reported in this work, we have used a copper polycrystal which was cut in the shape of a cylinder of diameter 2 mm and height 2.8 mm. The expansion measurement performed with the reference copper sample yields the empty cell effect, $\Delta L_{\text{emptycell}}$, which is the difference of the cell length change $\Delta L_{\text{cell}}$ from the reported literature values of length change of copper, $\Delta L_{\text{Cu literature}}$:

$$
\Delta L_{\text{Cu}} = \Delta L_{\text{emptycell}} = \Delta L_{\text{Cu literature}} - \Delta L_{\text{cell}}
$$

The above equation comes from the fact that the only part of the cell frame which contributes to the measured length change is a piece of the frame which has exactly the same length as the sample being measured. Therefore, the relative length change of any sample, $\Delta L_{\text{sample}}$, is the sum of the measured length change of the sample, $\Delta L_{\text{meas}}$, with the length change corresponding to the calibrated cell:

$$
\Delta L_{\text{sample}} = \Delta L_{\text{meas}} + \Delta L_{\text{cell}} = \Delta L_{\text{meas}} + \Delta L_{\text{emptycell}} + \Delta L_{\text{Cu literature}}
$$

The relative length change of the sample normalised to its initial length $L_0$ is, then, given by

$$
\left( \frac{\Delta L}{L_0} \right)_{\text{sample}} = \frac{\Delta L_{\text{meas}}}{L_0} - \Delta L_{\text{emptycell}} + \left( \frac{\Delta L}{L} \right)_{\text{Cu literature}}
$$

Figure 7(a) shows the length change of the cell $(\Delta L/L_0)_{\text{cell}}$ for dilatometer #1 on CCR #1 (green curve), dilatometer #2 on CCR #1 (red curve) and dilatometer #2 on CCR #2 (blue curve). Unfortunately, CCR #1 broke
down before we could measure dilatometer\#2 on it, so we do not have this data with us presently and is the subject matter for future work. The literature values of the relative length change ($\Delta L/L^0_{\text{literature}}$) and the corresponding coefficient of thermal expansion, $\alpha$, for a pure copper sample was taken from the reference [29].

Figure 7 (b) shows the calculated values of the relative length change corresponding to empty cell effect, ($\Delta L/L_0^{\text{empty cell}}$), using the equation (10). The colour code represents the same configuration as described in figure 7(a). The first observation to be made from the data plotted in figure 7(a) is that the data quality is very good, similar to the copper reference data (black curve) measured on a wet cryostat using liquid helium as a cryogen [29]. This observation, then, suggests that we have been able to mitigate the noise problems arising in dry cryogen free cryostats to such a level that the resultant data quality is very similar to that obtained from wet cryostats. This is very heartening in the light of the fact that liquid helium is a rapidly depleting cryogen, and, hence, one can reduce one’s dependence on it for doing extremely sensitive experiments like thermal expansion requiring sensitivity of the order of $\Delta L/L < 10^{-9}$. From figure 7(a), it can also be observed that ($\Delta L/L_0^{\text{cell}}$) is the same for both the dilatometers in both the cryostats till ∼90 K above which they differ in magnitudes. However, the temperature variation is smooth and monotonic for both. The data for dilatometer \#2 was found to be independent of the cryostat used since the two curves corresponding to CCR\#1 (red) and CCR\#2 (blue) were found to overlap.

Küchler et al [34] found a temperature independent empty cell effect ($\Delta L/L_0^{\text{empty cell}}$) till ∼200 K on a scale of 0.5·10$^{-3}$ when measured in a wet PPMS/exchange gas cryostat. If we compare our ($\Delta L/L_0^{\text{empty cell}}$) data shown in figure 7(b), on Küchler et al’s scale [34], the empty cell effect is nearly temperature independent till ∼280 K for dilatometer\#1 measured in CCR\#1. In comparison, for the dilatometer\#2 the temperature independence is over a smaller temperature interval of ∼150 K, comparable to what was obtained by Küchler et al [34] in their wet cryostat.

### 3.3. Thermal expansion of Aluminium

To check the functioning of the dilatometers, it is necessary to first calibrate it with other metals having well known thermal expansion characteristics. To do this, we performed thermal expansion measurements on an aluminium polycrystal which was cut into the shape of a cylinder of diameter 2 mm and height 3 mm for dilatometer\#1 and diameter 2 mm and height 2.57 mm for dilatometer\#2.

The values of the relative change in length $\Delta L/L$ measured with dilatometer\#2 using CCR\#1 and CCR\#2 are shown by red curves in figure 8(a) and (b) respectively, while figure 8(c) shows the same data with dilatometer\#1 using CCR\#1. Black continuous curve in each figure is the literature data taken from reference [29]. It can be seen that while the $\Delta L/L$ curve obtained with dilatometer\#1 using CCR\#1 (see figure 8(c)) shows a slight deviation from the literature curve ~140 K, the curves obtained with dilatometer\#2 using either of the CCR’s (see figures 8(a) and (b)) show an excellent match with the reference curve. Since the slight bump in $\Delta L/L$ values in the aluminium sample is observed on the same aluminium sample but only on dilatometer\#2 but not when measuring on dilatometer\#2 using either CCR\#1 or CCR\#2, it is clear that the bump is not intrinsic to the sample but is an artifact of the measurement procedure using dilatometer\#1.

Coefficient of thermal expansion $\alpha$ obtained for aluminium for the three different setups, as described above, are shown in figure 8(d) (green, red and blue), while the literature values of $\alpha$ obtained from reference [29] is shown in the black curve of figure 8(d), $\alpha$ was calculated using a polynomial fit, where a smooth polynomial fit was done on the quantities $\Delta L_{\text{meas}}$ and $\Delta L_{\text{empty cell}}$ prior to calculation of derivatives. The goal was to find coefficients $\beta$’s corresponding to a polynomial, $P$ of the type $P = \beta_0 + \beta_1 T + \beta_2 T^2 + \beta_3 T^3 + \ldots + \beta_n T^n$. This was achieved via a least square fit to the dataset by dividing the entire data range in sets of $m$ data points, so as to obtain $N/m$ different polynomials for $N$ total number of observations. For the calculation of $\alpha$ for aluminium, we took a set of 6 to 10 data points $(m \in \{6, 10\})$ and fitted the $N/m$ set of points to a polynomial of degree $\leq 3$. The resultant values are displayed in figure 8(d). As is evident from the graph, the calculated values of $\alpha$ for aluminium using dilatometer\#2 on either of the CCR’s have an excellent match with the literature values obtained on aluminium metal using a wet cryostat, thus proving the effectiveness of the CCR’s in obtaining sub-angstrom resolution thermal expansion values. Since the results for aluminium employing dilatometer\#1 is not good, it leads us to conclude that sub-angstrom resolution thermal-expansion measurements are not compatible with dilatometer\#1 in any CCR.

Our first thought on obtaining this result was a possible problem with our assembly of dilatometer\#1. However, repeated disassembly of the dilatometer and reassembly always gave us results of figure 6, thus ensuring that the dilatometer was working fine in the parallel limit. So, we believe that the reason for the existence of a slight bump in the $\Delta L/L$ curve in figure 8(d) is the design of the dilatometer\#1 itself. From 2(a), it can be seen that the spring $k$ is connected to the lower capacitor plate $g$ via the three $m$ screws located at 120° with respect to each other (non-constrained design of the spring). These screws are made up of soft Be-Cu alloy and screw the spring by the application of a small force. Too much force results in either smoothening up of the screw threads or shorting of the shims. So, when a small vibration from the CCR reaches the dilatometer (even though the vibrations in the CCR have been mitigated quite a lot by the design discussed above, small vibrations may still be reaching the CCR), it may result in a slight offset of the spring held by the not-so-tight screws, which...
results in a very slight tilt of the lower movable capacitor plate towards the upper capacitor plate. This results in an extremely slight deviation of the $\Delta L$ values that are picked-up by the very sensitive capacitance bridge. Such a problem does not arise in dilatometer $\#2$ since the spring $K$ in dilatometer $\#2$ is machined as part of the main dilatometer body, thus, constraining it’s motion. We have performed a FEM simulation (see below) to second this argument.

### 3.4. Thermal expansion of silver

From the results obtained for thermal expansion measurements performed on aluminium on the two dilatometers dilatometer $\#1$ and dilatometer $\#2$, and two different closed cycle refrigerators CCR $\#1$ and CCR $\#2$ (see figure 8) (d), we can clearly conclude that the dilatometer of choice for a thermal expansion measurement on a closed cycle cryostat is dilatometer $\#2$ (Küchler’s dilatometer) and either of the closed cycle refrigerators, CCR $\#1$-Gifford-McMahon based PPMS-Evercool or CCR $\#2$-Pulse tube based PPMS Dynacool. As an additional test of the good performance of dilatometer $\#2$ on CCR for thermal expansion measurements, we measured another metal with well known thermal expansion characteristics, namely, silver. Red continuous curve in figure 9(a) shows the plot of $\Delta L/L$ obtained on a polycrystal of silver using dilatometer $\#2$ and measured on CCR $\#2$, while the black filled circles correspond to literature values extracted from [34]. From the plot, it is clear that our obtained values of $\Delta L/L$ match the literature data very well. Figure 9 (b) shows the calculated linear thermal expansion coefficient $\alpha$ from the corresponding $\Delta L/L$ values of figure 9(a) obtained by the polynomial fitting algorithm described above. It can be seen that the experimentally obtained $\alpha$ matches the literature data very well in the entire temperature range, thus demonstrating the utility of dilatometer $\#2$ in performing sub-angstrom thermal-expansion measurements in closed cycle cryostats.

### 3.5. FEM simulation of uniaxial force exerted on a sample by the dilatometers

Since the dilatometer $\#2$ gave excellent data in both CCR $\#1$ as well as CCR $\#2$ as compared to dilatometer $\#1$, we wanted to check if the reason for the difference was a better noise mitigation in dilatometer $\#2$ as compared to dilatometer $\#1$. It is clear that the incorporation of the new experimental design led to a much better noise...
mitigation as compared to the first design (see Experimental setup section above). However, some very small magnitude noise could still creep in. To verify this, we performed finite element method (FEM) simulations on the spring movement (spring $k$ in dilatometer#1 and spring $K$ in dilatometer#2) of both the dilatometers under the action of a force/pressure. This force may arise either due to the expansion of the sample itself or due to noise vibration that may be imparted to the dilatometer spring. Since the magnitude of the forces applied are very small, it may result in small non-monotonicities in the thermal expansion coefficient data, which may affect the measurements artificially. Additionally, the sensitivity of the dilatometers is critically dependent on the thickness of the springs used in them. However, machining a dilatometer spring of a given thickness, assembling it in the dilatometer and then measuring thermal expansion is an extremely tedious process. So, in order to check if a monotonic variation of displacement is achieved on application of forces with varying magnitudes as well as obtain thickness variation of displacement of a given dilatometer spring, we performed FEM simulations on various digital models of the spring, having a geometry similar to that used in dilatometer#1 [31] as well as dilatometer#2 [34]. The simulation was done using COMSOL Multiphysics simulation software package which is a software cross-platform for finite element analysis, equation solving and multi-physics simulation [52]. To build the various models of the circular spring [31] needed as an input to the COMSOL software, we used CREO Parametric software. For a particular model, the displacement field at various points was obtained by (i) uploading the model with desired thickness and diameter, (ii) various parametric curves drawn to divide the surface into regions, (iii) specifying the boundary condition and boundary load, and (iv) choosing a mesh size depending on the precision of the measurement—finer the mesh, more accurate the results.

3.5.1. Dilatometer #1

We started off by finding the displacement of the spring under the application of pressure (force per unit area), with values chosen in the range that produce displacements in the angstrom range. The application of a pressure results in von Mises stress in different parts of the spring [33]. It was found that application of large pressures of the order of
1000 N/m² resulted in the spring getting deformed. However, in the low range of applied pressure of the order of 0.05 N/m² - 0.40 N/m², resulting in displacements of the order of few angstroms, the spring was well within the elastic limit, causing only elastic deformations. Figure 10 (a) shows a representative image of the deformation caused on the dilatometer spring in the z direction as well as the variation of the von Mises stress on the application of a pressure of magnitude 0.2 N/m². The material input for the simulation was beryllium-copper (Be-Cu) alloy UNSC17200. This is the same alloy that we have used in fabricating our dilatometer #1. Schmiedeshoff et al [31] have also used the same material in their dilatometer. A single hole at the centre of the spring represents the hole made in the spring for holding the lower capacitor plate via the nut (refer to figure 2 (a)). The holes at the circumference of the circular spring correspond to the holes made in the spring to screw the spring to the cell frame a by three m screws, shown upside down in figure 2 (a). For the FEM simulation, this region is constrained to be fixed and is one of the boundary conditions that was applied, i.e. the applied pressure is zero at the position of the holes. The other boundary condition is that the maximum pressure is applied at the central region of the spring via a contact force that gets applied on the spring via the lower capacitor plate when the sample expands. Accordingly, the area of the holes at the circumference of the spring is shown as dark blue region and the central hole region is shown as dark red in figure 10 (a). The region between these two colours (dark blue and dark red) shows the distribution of von Mises stress on application of a pressure in the central region.

The mesh size for the FEM calculation was dynamically allocated depending on the curvature of the surface, namely, a fine mesh was chosen near the holes and the edges, while a coarse mesh was chosen at other places. Each element of the FEM was chosen as a tetrahedron with a minimum element size of 0.686 mm and a maximum element size of 3.814 mm. The x and y co-ordinates in figures 10 (a) and (b) denote the spring size in mm to represent the diameter of the Be-Cu spring which is ~19 mm. The z co-ordinate in figure 10 (a) is in the scale of 10⁻⁷ mm, to reflect the displacement of the spring that occurred in the spring on being subjected to forces via the sample. Figure 11 (a) shows the displacement of the spring that was subjected to pressures described in figure 10(a) above. The resultant displacement, plotted as a function of the radial distance, occurs as a result of the application of the pressure in the basal plane of the spring. It can be seen that the displacement is the maximum at 1.5 mm from the centre of the spring corresponding to the area where the pressure is applied. As one moves away from the centre, the displacement decreases.
decreases and becomes ∼zero at 6 mm. This is the position where the nuts are placed. From figure 11(a), it is clear that the displacement varies linearly with radial distance in the range 1.5 mm to 5 mm for all values of applied pressures. Figure 11(b) shows the variation of the obtained displacement with respect to the applied force. It can be seen that the variation is clearly linear which is confirmed by a straight line fit, shown as red line in figure 11(b). The slope of the fit gave the spring constant as 38 843 N/m, in agreement with values obtained on springs made of similar material [34], implying that our simulations are correct.

In order to see the effect of thickness of the spring on a resultant displacement, we simulated a displacement versus radial distance curve for varying thicknesses of the spring, starting with 0.227 mm and ending with 0.077 mm. Figure 11(c) plots this curve where it is very clear that the minimum displacement obtained at a given radial distance is the minimum for the thickest spring of 0.227 mm while the maximum displacement is obtained for the thinnest spring of 0.077 mm. However, it is also to be noted that the displacements obtained by the 0.102 mm spring is also quite large. We operate our dilatometer with this thickness of 0.1 mm. A lower thickness spring can give higher displacements but is extremely difficult to machine and may also result in easy deformations, so we fixed our thickness to 0.1 mm.

It was found that the dilatometer #1 gave an artificial bump in the $\Delta L/L$ values and a corresponding mismatch of $\alpha$ for aluminium when compared to the literature data (see figure 8(d) and discussions therein). To explain the mismatch, we postulated that this artifact may be arising due to a slight displacement of the dilatometer #1 spring resulting in a slight tilt of the lower capacitor plate itself. In order to confirm this, we applied a pressure at a radially asymmetric point of the dilatometer #1 arising from a sudden vibration from the cold head of either of the CCR. Figure 10(c) shows the result of such a simulation where the pressure is maximum at a point A where the von Mises stress is the maximum (shown by red colour). It can be clearly seen from figure 10(c) that the displacement of the spring is not symmetric in the z-direction near this area where the pressure has been applied, resulting in a slight tilt of the spring around this area. Since the spring is connected to the movable lower capacitor plate via the screws, it results in a slight tilt of the lower capacitor plate resulting in non-parallelness of the two capacitor plates, generating an artificial variation of $\alpha$.

Figure 11. (a) Variation of displacement of the spring of thickness 0.127 mm along the radial direction in the basal plane as a function of different values of pressures applied over an area of 23.127 mm$^2$. (b) Maximum displacement (co-ordinate of the center of the spring) versus the applied force on the sample. Black dots represent the data from graph (a) and the red line represents a linear fit to the data. The slope of the graph gave the spring constant as 38 843 N/m. (c) Deformation of the BiCu springs of thickness 0.077 mm (black), 0.102 mm mm (red), 0.127 mm (green), 0.177 mm (blue), 0.227 mm (black) under same applied force of 1N.
3.5.2. Dilatometer #2

If a small vibration from the CCR resulted in a slight displacement of the spring in dilatometer #1 as seen in the simulation above, then it would result in an asymmetric force on the spring in dilatometer #1, affecting the measurements artificially. Since such an observation was not made in dilatometer #2, it is clear that the design of dilatometer #2 is better suited to ward off possible noise vibrations arising from the CCR. As described in the ‘Experimental setup’ section above, dilatometer #2 was machined out of a single block of BeCu and contained the main cell body, the two capacitor plates and the leaf springs. The mechanical constraint placed by such a design of the spring constrains the movable capacitor plate to move in only one direction offering very high parallelism between the two capacitor plates [27, 32, 34]. In order to demonstrate this, we have performed FEM simulations on a model of the dilatometer #2. The input drawings for the simulations were made using CREO Parametric software. The material chosen for the simulation was the same Beryllium–copper (Be-Cu) alloy as specified by Schmiedeshoff et al [31], namely, UNSC17200. The first objective of the simulation was to show the parallel movement of the spring even under the application of a force applied at an asymmetric point. An asymmetric force can be applied onto the dilatometer spring from vibrations imparted to the dilatometer spring from a closed cycle cryostat. So, the first boundary condition of the simulation is maximum force application at point P (shown in figure 12(d)). The second boundary condition is zero force at the left blue shaded area of figures 12(a) and (b). Parallel movement of the spring was found at the application of very small forces of the order of few μN as well as large values of forces. Since the capacitor plates move parallel to each other under the application of a force at an asymmetric point, it confirms our hypothesis that the reason for the smooth variation of the thermal expansion in either of the CCR’s using dilatometer #2 lies in the parallel motion of the capacitor plates of dilatometer #2 even if it is subjected to a vibration from any of the CCR’s.

For calculation of the spring constant, the value of the force was chosen to be in conformity to the values of forces that were applied by Küchler et al [34] in their actual measurement of the relative change in length brought about by the application of a force in dilatometer #2. The resultant variation of von Mises stresses on the leaf spring due to the application of this pressure is shown in figure 12(a). The mesh that were chosen for the entire flange-spring unibody system to execute the FEM simulation were dynamically allocated and are shown in figure 12(b). Each element of the mesh was chosen as a tetrahedron with the size of the largest element chosen at the cell body as 3.814 mm while at the leaf spring the finest mesh was chosen as 0.686 mm. The simulation was performed using a time dependent solver in the COMSOL software, where a fixed constraint was applied to the
part opposite to point P. Figure 12 (c) shows the cross-sectional view of the resultant displacement of the leaf spring-flange unibody system. It can be clearly seen from the figure that the application of an asymmetric force at a corner of the cell body results in a parallel movement of the leaf spring-flange unibody without any bend. Figure 12 (d) shows the time evolution of the obtained displacement in μm as a function of the distance along the x-axis. It is to be noted that our obtained values of displacement are in the same order of magnitude (μm) as that obtained in [34]. The simulation was performed for a time of 0.2 s. As can be seen from figure 12(d), the displacement of the spring-flange unibody is maximum at the point of application of the force (~6 mm away from the centre of the x-axis) and goes to zero at ~−6 mm from the centre of the x-axis. The values of the maximum displacement obtained at the point P increases as a function of time, as expected.

In order to see the effect of the variation of applied force on the displacement of the leaf spring, we simulate displacement versus distance curves as shown in figure 13 (a). The values of applied forces ranged from 0.09 N (dark blue) to 1.47 N (black). From figure 13(a), it can be clearly seen that the displacement rises from zero value at ~−6 mm away from the centre of the x-axis and reaches a maximum at +6 mm. The value of obtained displacement at a given position along the x-axis also increases with an increase in the value of applied force, in exact conformity with the results obtained for dilatometer #1 (cf. Figure 11 (a)). Red open circles in figure 13(b) represent the maximum value of displacement for a given value of applied force obtained from figure 13(a). The values of applied forces were chosen to be of similar values as the one used by Küchler et al in their force versus displacement curve (cf. Figure 4 of [34]). Black open circles in figure 13(b) denote the extracted values of displacement from reference [34]. It can be seen that our simulated values match those obtained from [34] very well and the variation of force with displacement is linear. A straight line fit to the data, shown as green solid line in figure 13(b), gave the slope as 24 530 which corresponds to the spring constant of the material. The obtained value of the spring constant is in excellent agreement with the value 24 291 N/m obtained by Küchler et al [34].

3.6. Thermal expansion measurements on YBa2Cu3−xAlO6+x and Bi2Sr2CaCu2O8+x

After describing the details of the two closed-cycle cryostats and the two dilatometer’s functioning on standard metal samples, we measured thermal expansion on single crystals of two different high temperature superconductors, namely, YBa2Cu3−xAlO6+x (Al-YBCO) and Bi2Sr2CaCu2O8+x (BSCCO-2212). The measurements were done on dilatometer #2 on CCR #2, i.e. on Küchler’s dilatometer and dynacool closed cycle refrigerator. The single crystal of Al-YBCO selected for the measurements was an as-grown single crystal that was grown by a self-flux method employing a vertical temperature gradient and had a superconducting onset temperature $T_{c onset} = 58.5$ K [18]. The as-grown crystal was not twinned and no detwinning was applied on the crystals before the thermal expansion measurements in order to avoid any defect incorporation in the crystals [12]. The length of the crystal along the c-axis is 1.02 mm.

Red filled circles in figure 14 denote $\Delta L/L$ values obtained on the single crystal of Al-YBCO measured along the c-axis while the blue filled circles denote the corresponding thermal expansion, $\alpha$. The data was obtained by following the polynomial fit method described above. So, a second order polynomial was chosen for the fit with the number of data points fitted simultaneously, $N = 60$. The error on the calculated $\alpha$ amounts to the error on the temperature, $\Delta T$ and the measured capacitance $C$. The experimental error on the temperature, $\Delta T$ was found to be very small, of the order of 1 mK. The error on capacitance, $\Delta C$, is found to be 0.15%. So, the error $\Delta \alpha$, on thermal expansion is ~5%. From figure 14, it can be seen that the $\alpha$ curve shows a smooth variation with
temperature, in agreement to other reports of temperature variation of \( \alpha \) measured along the \( c \)-axis in Al-YBCO \([11, 12, 15, 28]\). For comparison, we have plotted the measured thermal expansion coefficient \( \alpha \) obtained on the same Al-YBCO single crystal by using the polynomial fit on \( \Delta L/L \) data. Error bars on the data denote the error on \( \alpha \) that was obtained from errors in measured capacitance and temperature values. See text for details. Blue labels on the right represent the corresponding \( \alpha \) values. Onset of superconducting transition temperature \( T_{c}^{\text{on}} \) is marked by a vertical arrow. Open blue circles represent literature values of \( \alpha \) extracted from \([11]\).

Figure 14. Red filled circles denote \( \Delta L/L \) values obtained on an as-grown single crystal of YBa\(_2\)Cu\(_{3-x}\)Al\(_x\)O\(_{6+\delta}\) (Al-YBCO) measured along the \( c \)-axis. Red labels on the left represent the corresponding \( \Delta L/L \) values. Filled blue circles denote the linear thermal expansion coefficient \( \alpha \) obtained on the same Al-YBCO single crystal by using the polynomial fit on \( \Delta L/L \) data. Error bars on the data denote the error on \( \alpha \) that was obtained from errors in measured capacitance and temperature values. See text for details. Blue labels on the right represent the corresponding \( \alpha \) values. Onset of superconducting transition temperature \( T_{c}^{\text{on}} \) is marked by a vertical arrow. Open blue circles represent literature values of \( \alpha \) extracted from \([11]\).

Figure 15. Measured values of \( \Delta L/L \) obtained on Bi\(_2\)Sr\(_2\)Ca\(_2\)Cu\(_3\)O\(_{8+x}\) superconductor shown in red colour. Linear thermal expansion coefficient \( \alpha \) obtained by using a polynomial fit on the corresponding \( \Delta L/L \) data is shown in blue filled circles. The labels on the right are the corresponding \( \alpha \) values. Error bars on the data arise from the errors in measured capacitance and temperature values as described in the main text above. Onset of superconducting transition temperature \( T_{c}^{\text{on}} \) is marked by a vertical arrow. Open blue circles represent literature values of \( \alpha \) extracted from \([13]\).

temperature, in agreement to other reports of temperature variation of \( \alpha \) measured along the \( c \)-axis in Al-YBCO \([11, 12, 15, 28]\). For comparison, we have plotted the measured thermal expansion in Al-YBCO along \( c \)-axis by extracting the data from \([11]\), which is displayed by open blue circles measured on a wet cryostat. It can be seen that our data quality is the same as that obtained in \([11]\). Since the high temperature superconductor’s properties vary dramatically with their oxygen content, homogeneity of the oxygen content etc., it is expected that our experimentally obtained curve would not exactly overlap with the literature curve. However, the point to be noted is that the quality of our data obtained using a CCR is similar to that obtained in the comparative literature plot obtained using a wet cryostat, proving the usefulness of our set-up for measuring sub-angstrom resolution thermal expansion using closed-cycle cryostats. It is also to be noted that the thermal expansion data measured along the \( c \)-direction of Al-YBCO does not give any jump at \( T_c \), but only a smooth curve, a very well known fact about Al-YBCO as well as BSCCO-2212 \([11–13, 15, 17, 28]\). Guided by the magnetisation data \([18]\), we have marked the onset temperature of superconductivity, \( T_c^{\text{on}} \), as 58.3 K.

Finally, we present thermal expansion data on the single crystal of another high temperature superconductor BSCCO-2212. The single crystal was grown by the self-flux regrowth method and had a \( T_c^{\text{on}} \) of 88.3 K. The details of the crystal growth and characterisation of its structural and superconducting properties can be found in...
The measurements were done for the length along the crystallographic c-axis and the chosen crystal had a total length of 1.23 mm along c-axis. Similar to the data on Al-YBCO, red filled circles in figure 15 denote the ΔL/L values for BSCCO-2212 crystal while the blue filled circles denote the linear thermal expansion coefficient α extracted from the corresponding ΔL/L values. The values were obtained with a polynomial fit of second order and N = 60. Our values of ΔL/L and α match quite well with other reported values [13, 17]. For comparison, we have plotted the extracted values of α from [13] measured on a single crystal of BSCCO-2212 as open blue circles. It can again be noted that our data quality matches very well with that in [13] obtained using a wet cryostat. Similar to the observation in Al-YBCO, the linear thermal expansion coefficient α is found to vary smoothly with temperature without any discontinuities [13, 17]. We have marked the onset temperature of superconductivity Tc as 88.3 K, found from magnetisation measurements [54].

4. Conclusion

To conclude, we have demonstrated the utility of closed-cycle cryostats to perform thermal expansion measurements which are known to be extremely sensitive to noise vibrations. We employed two different kinds of dilatometers for the thermal expansion measurements, one that was built in-house and the other that was procured commercially. We tested the functionality of the two dilatometers on two commercially available closed cycle cryostats one of which works on the principle of Gifford-McMahon cooling while the second employs pulse-tube cooling technology for achieving the closed-cycle refrigeration. Details of the experimental set-up utilising a multi-function probe integrated with a closed-cycle refrigerator has been described. By decoupling the bottom sample puck from the cryostat and taking connections from the top of the multifunction probe, smooth and continuous capacitance data could be obtained. By a series of thermal expansion measurements performed on polycrystals of standard metals, aluminium and silver, we demonstrate excellent data, similar to those reported in wet cryostats. Linear thermal expansion α was obtained using derivative of a polynomial fit and found to be in excellent agreement to those obtained using wet liquid helium based cryostats. It was also found that the dilatometer based on Schmiedeshoff’s design (dilatometer#1) [31] doesn’t give as good quality data as that obtained using Kühcher’s dilatometer (dilatometer#2). However, with dilatometer#2, very good quality data was obtained in either of the pulse tube based cryostat or Gifford-McMahon based cryostat. We, then, performed finite element method simulations on both the dilatometers to understand the spring action motion under the action of a force/pressure and vibrations on the spring motion. This was done to find if a parallel movement of capacitor plates is reached even under the application of force on an asymmetric point. For the dilatometer#2, this was found to be the case and we speculate that it helps in mitigating the vibrational effects of the cold head of the cryostat in dilatometer#2. We were also able to propose optimal thicknesses of dilatometer springs in both dilatometer#1 and dilatometer#2 for maximal displacement. Finally, we measured thermal expansion on single crystals of two high temperature superconductors, YBa2Cu3−xAlxO6+δ and Bi2Sr2CaCu2O8+δ along the crystallographic c-axis and found very good match with published data obtained using wet cryostat, demonstrating great technological possibilities for future thermal expansion measurements using closed cycle cryostats.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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