A miniature capacitance dilatometer for thermal expansion and magnetostriction

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A very small capacitive sensor for measuring thermal expansion and magnetostriction of small and irregular shaped samples has been developed. A capacitive method with tilted plates is used. The tilted plate capacitance formula is used for the calculation of the capacitor gap, the calibration is performed by measuring the signal of a standard material. The active length of the sample can be less than 1 mm. The absolute resolution is about 1 Å. All mechanical connections of the dilatometer are carried out by tiny Cu–Be springs, enabling the small force on the sample to be adjusted (50–500 mN) and no additional sample fixing is necessary. The cell has been tested in the temperature range 0.3–200 K and in static magnetic fields up to 15 T. The zero signal of the dilatometer has been determined by measuring a silver sample. The correct operation and reproducibility has been verified by measuring the thermal expansion of Cu. The thermal expansion and magnetostriction of a $DyCu_2$ single crystal has been determined. The advantage of this method compared to specific heat measurements is that a large temperature range can be covered with one equipment. This high static and dynamic range of sample length, temperature, and magnetic field suggests a number of possible applications, like the investigation of crystal field effects on the magnetoelastic properties of single crystals or structural phase transitions. © *1998 American Institute of Physics.* [S0034-6748(98)01007-7]

I. INTRODUCTION

The capacitance method is one of the most sensitive methods for measuring small length changes of solids. In practice the accuracy is limited frequently by mechanical sample quality and dilatometer effects. However, to reach the highest possible sensitivity and reproduceability, special effort is needed in the design of the capacity cell and the sample preparation. Therefore a dilatometer has been developed by combining our own experience with already published methods.

The roots of capacitance dilatometry date back to 1961, when White[1] combined experiences of a two-terminal capacitance method for measuring thermal expansion[2] with Thompson's[3] three-terminal method for capacity measurements using a ratio transformer bridge. He achieved a hitherto unreachable resolution of 10^{-7} mm. Afterwards the two-terminal method was scarcely used again.[4]

White's design principles of absolute and relative dilatometers were adopted and improved by a number of different authors. It led to absolute thermal expansion measurements on a number of reference metals[5] like Cu,[6–8] Ag,[7] Au,[7] and Al.[8] Green,[9] Chanddrasekhar, and Fawcett[10,11] were among the first to use the dilatometers for magnetostriction measurements. Tilford and Swenson used an inverted configuration of White's cell to measure the thermal expansion of solid Ar, Kr, and Xe.[12] Miller *et al.* extended the temperature range of White's cell (1–300 K) up to 550 K.[13] The reproducibility of the dilatometer was increased by replacing the oxygen-free copper reference rods with silicon[14] or sapphire.[15] However, using for the whole cells Si or quartz with metal-plated electrodes[16,17] is problematic and has not been widely adopted except in pulsed magnetic fields.[18] Subrachmanyam and Subramanyam[19] went back to a copper dilatometer for the use of samples differing in length, however, still samples should have parallel surfaces. Sparavigna *et al.* designed an apparatus for simultaneous measurement of thermal expansion and thermal diffusivity[20] still keeping the main features of White's dilatometer design.

Another method—differing from White's design—goes back to the pushrod dilatometers with capacitive displacement sensor kept at constant temperature.[21,22] However, the pushrod has been used mainly at temperatures above room temperature.[23,24] The idea of separating the sample from the displacement sensor was fascinating, because samples of different lengths and shapes can be used. Several authors applied this principle in dilatometers with a capacitor based on parallel spring movements[25] which is not dismantled on sample change.[26–30]

The drift of capacitance with time could be reduced by the use of sapphire isolation washers instead of epoxy–mylar isolation.[29,31] Studies of the absolute accuracy of capacitance displacement sensors[32–34] led to further developments, like the elastic diaphragm sensor.[35]

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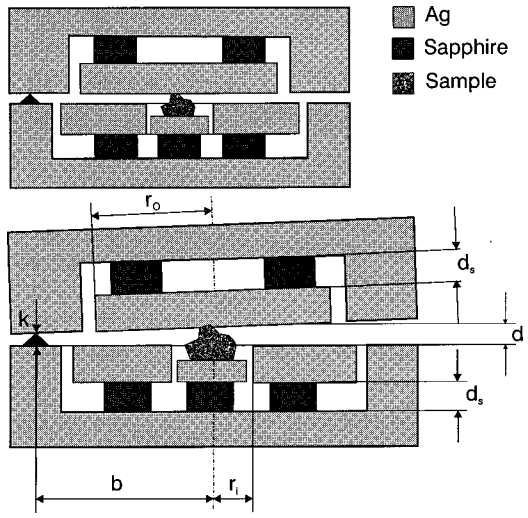

FIG. 1. Schematic drawing of the capacitance dilatometer.

An important basis for the design of our sensor was the invention of the tilted plate principle,[26,36] which has also been used in a dilatometer for amorphous ribbons.[37]

II. THE DILATOMETER

Our aim was to construct a sensor for studying phase transitions on intermetallic rare earth compounds. Since for these compounds only small single crystals ($1\ mm^3$) are available and for such investigations a wide range of physical parameters is necessary, it was required to design a small and compact dilatometer for a wide temperature range and high magnetic fields combining most advantages of the existing capacitive dilatometers but avoiding their disadvantages.

Capacitive cells with parallel plates are easy to calibrate, but they have either big dimensions or difficulties with the sample handling. Even if the problem of thermal stability of such big cells is solved, limited space in most of the magnetic coil systems causes problems, particularly when magnetostriction parallel and perpendicular ($\lambda_\parallel$ and $\lambda_\perp$) to the field has to be measured. To minimize the cell size our capacitor design is based on the tilted plate principle[26,36] with the sample placed in a hole in the lower capacitance plate (see Fig. 2), determining the maximal sample size ($3\times3\times3\ mm^3$). To obtain a reasonable accuracy the active length of the sample should be bigger than 0.5 mm. The sample can have nearly any irregular shape, only the base surface should be flat to give a stable sample position.

Our tilted plate construction has a high plate area and a low volume, which gives a good sensitivity in connection with very small sensor dimensions (diameter: 22 mm, height: 14 mm).

Figure 1 shows the schematic arrangement of the dilatometer. The lower part consists of a plate holder (Ag). It includes the ringlike lower capacitor plate (Ag) and the sample support (Ag). The upper part consists of the upper plate holder (Ag) and the disklike upper capacitor plate. It is separated from the lower one by two needle bearings (brass) and the sample to obtain a well defined support on three points. Both capacitor plates as well as the sample support

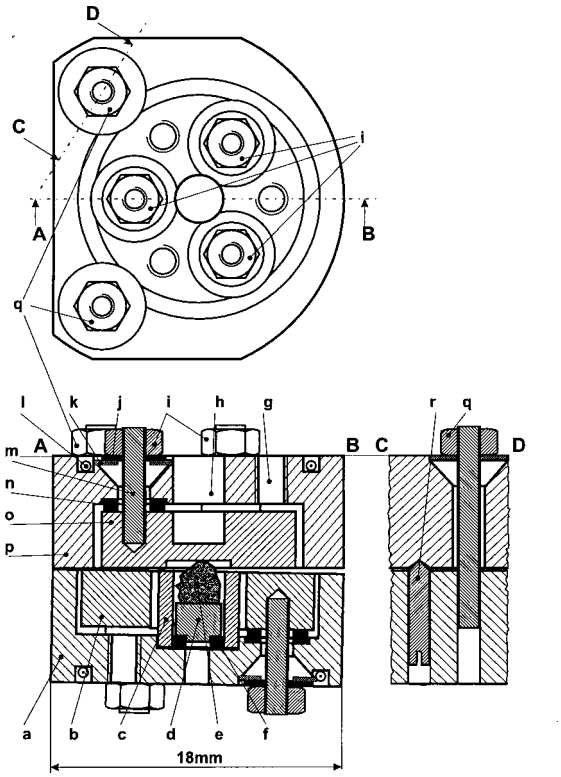

FIG. 2. Detailed drawing of the miniature tilted plate capacitance dilatometer: (a) lower capacitance-plate-holder (Ag), (b) lower capacitance-plate (Ag), (c) electrical shielding of sample space (brass), (d) sample support (Ag), (e) sample, (f) isolation washer for electrical sample isolation (sapphire), (g) mounting holes, (h) temperature sensor hole, (i) plate holder nuts (brass), (j) electrical isolation (kapton), (k) disk spring (Cu–Be), (l) groove for temperature stabilization of capacitance wires, (m) thread bolt (brass), (n) isolation washer for electrical isolation of capacitance plate (sapphire), (o) upper capacitance plate (Ag), (p) upper capacitance plate holder (Ag), (q) adjustment nuts, and (r) needle bearing.

are insulated from the holders by sapphire washers.[29] The needle bearings define an exact pivot point and avoid any transversal shift between the upper and lower plate holder.

Figure 2 shows the detailed drawing of the complete dilatometer. In addition to the schematic drawings the following parts are shown. The electrical shielding of the sample (c) is essential for screening the sample support (d) from the lower plate. The groove (l) holds the capacitance wires and ensures a good thermal contact to improve temperature stabilization. The thread bolt (m) together with the disk spring (k) fix the position of the capacity plate and performs the electrical connection to the plate. The disk spring fitting in the conical hole produces a well defined force to keep the position of the capacity plate with respect to the holder.[29,31] This improved design looks much simpler than that one of Pott and Schefzyk.[29] The stress on the sample can be adjusted with a torque driver at the nuts (q). To obtain a good electrical insulation between plate holders [(a) and (p)] and the capacitor plates [(b) and (o)] the kapton insulation ring (j) and the Cu–Be disk spring (k) must be aligned exactly and fit precisely in the hole.

Although this sensor is more difficult to calibrate than normal parallel plate dilatometers, several advantages outnumber this computational effort:

(1) A self-compensating construction gives a small temperature dependence of the zero signal. Sample support and lower capacitance plate are placed on sapphire washers of the same kind to perform equivalent motions during expansion (see Fig. 1).
(2) The inverting construction[12] causes that the expansion of the sample opens the capacitor. This provides a high dynamic measurement range and avoids crashing of the capacitor plates and sample damage.
(3) All mechanical connections are done by Cu–Be disk springs, which are pressed into a cone by an adjustment nut. In this way the following features are obtained:
—the stress on the sample can be adjusted,
—no other fixing of the sample is necessary in magnetic fields, (i.e., by glues which might get loose at low temperatures and high fields),
—all stresses are well defined on assembling the cell (using a special torque driver).
(4) The insulation of the capacitance plates is done by using sapphire washers.[29,31] This avoids the use of glue, which causes excessive (undefined) capacitance drift.
(5) The sample is placed in the center of the capacitor and the capacitance leads are thermally anchored to the cell in a special *V* slot. This gives excellent thermal equilibrium conditions of the sample and all other parts of the dilatometer.
(6) The essential parts of the dilatometer are made of silver,[38] this has the following advantages compared to OFHC copper;
—50% lower heat capacity per volume,
—no nuclear heat capacity at low temperatures and high magnetic fields.
(7) Simple construction (four parts and a set of sample supports) facilitates manufacturing and cleaning.
(8) To measure samples of different lengths and shapes a set of sample supports with several lengths is used. This avoids the adjustment by a sample screw, which introduces an additional source of uncertainty.
(9) The dilatometer range can easily be extended to temperatures above room temperature. Because of the used insulation materials (sapphire, kapton) an application of the cell up to 500 K seems to be possible without major changes, but has to be verified by further experiments. Choosing other materials the use of the design up to 1300 K and more is suggested. The extension of the temperature range of the dilatometer below 300 mK should also be possible.

Figure 3 shows the dilatometer in a temperature insert of the cryostat and in a superconducting magnet system. The two different positions in the coil parallel and perpendicular to the magnetic field are outlined as full and dashed lines, respectively.

III. CALIBRATION

This sensor measures the relative length change of the sample, therefore a calibration procedure is necessary.[1] It is performed using the tilted plate capacitance formula.[26,36]

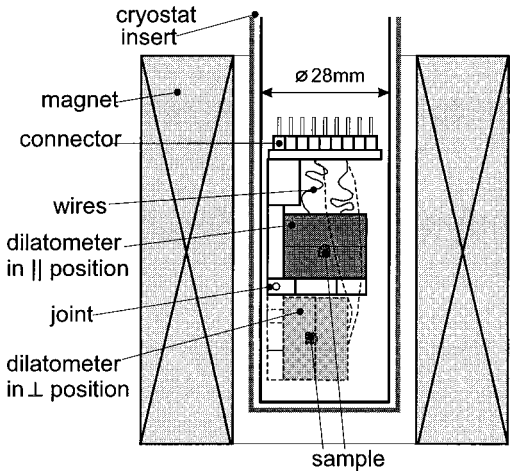

FIG. 3. Arrangement of the dilatometer, temperature insert, and magnet system.

This presents no major problem using computer controlled data acquisition.
The capacity change caused by thermal expansion has four contributions;

(1) change of the sample length,
(2) change of the radii of the capacitor plates,
(3) change of the length of the silver plate holders,
(4) partly compensated by the change of thickness of the capacitor plates and the sapphire washers.

The measured capacity $C(T)$ is used to calculate the gap $d(T)$ by the following formula:

$$C(T)=\frac{2\epsilon_0}{d(T)}\left[A_0(T)\,\frac{(1-\sqrt{1-\gamma_0^2})}{\gamma_0^2}-A_i(T)\,\frac{(1-\sqrt{1-\gamma_i^2})}{\gamma_i^2}\right], \tag{1}$$

with

$$\gamma_0=\frac{r_0}{b}\left[\frac{k(T)}{d(T)}-1\right], \tag{2a}$$

$$\gamma_i=\frac{r_i}{b}\left[\frac{k(T)}{d(T)}-1\right], \tag{2b}$$

where r_0 is the outer plate radius, r_i the inner plate radius, b the distance between center of capacitor and pivot (see Fig. 1).

$$k(T)=k(T_0)+2d_s\left[\frac{\Delta l_{\mathrm{Ag-Lit}}}{l}(T)-\frac{\Delta l_{\mathrm{Sapphire}}}{l}(T)\right], \tag{3}$$

where $k(T_0)$ is the pivot distance at $T_0=300$ K, d_s the thickness of sapphire washers (0.8 mm), $(\Delta l_{\mathrm{Ag-Lit}}/l)(T)$ is the thermal expansion of Ag from literature,[7] and $\Delta l_{\mathrm{Sapphire}}/l\,(T)$ the thermal expansion of sapphire from literature.[5]

To determine the pivot distance $k(T_0)$ the plates can initially be adjusted parallel (by minimizing the reading of the capacitance bridge and/or by the procedure described by Villar *et al.*[14]).

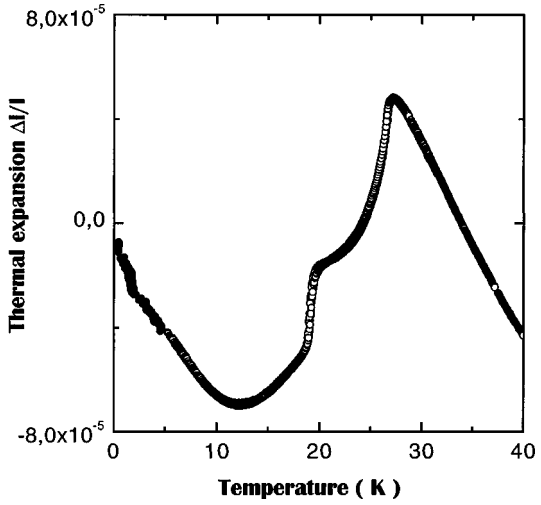

FIG. 4. Thermal expansion along the *a* direction of a $DyCu_2$ single crystal measured by the capacitive dilatometer.

$$A_i(T)=A_i(T_0)\left[1+\frac{\Delta l_{\mathrm{Ag-Lit}}}{l}(T)\right]^2 \tag{4a}$$

is the inner capacitance area ($r_i^2\pi$).

$$A_0(T)=A_0(T_0)\left[1+\frac{\Delta l_{\mathrm{Ag-Lit}}}{l}(T)\right]^2 \tag{4b}$$

is the outer capacitance area ($r_0^2\pi$).

Because $d(T)$ appears in the term of $C(T)$ [Eq. (1)] and in γ [Eqs. (2a) and (2b)], Eq. (1) has to be solved numerically with respect to $d(T)$.

Fringe corrections have to be considered only if big capacitance changes occur. For signals in the magnitude of a ''normal'' metal (like Ag or Cu) fringing corrections can be neglected. In this case an estimation according to W. C. Heerens[34] leads to corrections which are in the order of magnitude of the reproduceability of the cell. For big capacitance changes the corrections can be applied, therefore it is necessary to measure the capacity two times with inverted high and low connections.

Once the gap $d(T)$ has been calculated for each temperature, the thermal expansion of the sample $\Delta l_{\mathrm{Sample}}(T)/l$ is obtained by the following relations:

$$\frac{\Delta l_{\mathrm{Sample}}}{l}(T)=\frac{\Delta d_{\mathrm{Sample}}}{l_{\mathrm{Sample}}}(T)-\frac{\Delta d_{\mathrm{Ag-Sample}}}{l_{\mathrm{Ag-Sample}}}(T)+\frac{\Delta l_{\mathrm{Ag-Lit}}}{l}(T), \tag{5}$$

where $\Delta d=d(T)-d(T_0)$, $\Delta d_{\mathrm{Sample}}(T)$ is the measurement of the sample, and $\Delta d_{\mathrm{Ag-Sample}}(T)$ is the measurement of an Ag–Sample (calibration sample).

The capacity has been measured by an ''Andeen Hagerling 2500 A 1 kHz Ultraprecision Capacity Bridge.'' A gap of 0.18 mm results in a capacity of approximately 4 pF.

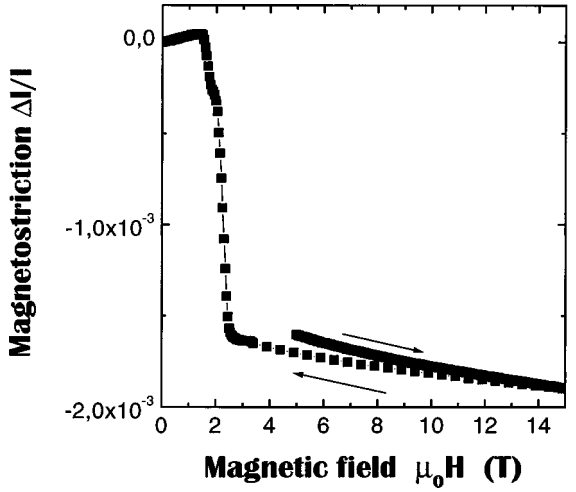

FIG. 5. Longitudinal magnetostriction of a $DyCu_2$ single crystal with the magnetic field aligned parallel to the *a* direction measured by the capacitive dilatometer at $T=4.3$ K.

IV. RESULTS

The correct operation has been verified by measuring the thermal expansion of pure copper (5 N). The maximum deviation from literature data[5] is in the order of 1% in $\Delta l/l$.

After calibration experiments the dilatometer performance was checked by extensive and systematic measurements on intermetallic compounds. Both temperature dependent runs at a constant magnetic field (up to 15 T) and field dependent runs at a constant temperature (from 300 mK to 200 K) have been performed. Above 2.2 K a variable temperature insert prepared for a high field magnet with 50 mm bore was used. Alternatively, the dilatometer was mounted in a ^{3}He insert with a sample space diameter of 40 mm for the low temperature experiments from 300 mK to 4 K. Results for a $DyCu_2$ single crystal with sample size 1.13 (a)×2.12 (b)×1.79 (c) mm^3 can be seen in Fig. 4. The compound shows two magnetic phase transitions (18 and 27 K)[39,40] connected with significant jumps in sample length. Several temperature cycles with different slopes (1–5 mK/s) have been performed above 2.2 K. The deviation of the different curves is within the symbol size, showing that the reproduceability of the dilatometer is in the order of 10^{-7} mm.

Figure 5 shows the magnetostriction of $DyCu_2$ for $T=4.3$ K. The resolution does not depend on the field value. The jumps at 1.5 and 2.0 T are connected with transitions between different magnetic states of the sample.

Eddy currents cause a additional heating of the cell, depending on the field sweep rate. The power supply of the magnet allows us to work with constant sweep rate of 0.2 T/min. At 0.8 K a constant warm up of 0. K which stays stable within 1 mK is observed. Above 2 K this effect can be neglected.

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