CAPACITANCE DILATOMETRY MOVES FROM CRYOGENIC TO HIGH TEMPERATURES

by M.O. Steinitz

A technique originally developed for low-temperature applications, capacitance dilatometry has recently become possible at elevated temperatures. The method is maturing and becoming easier due to developments in design and in electronics. Offering Angstrom resolution, this technique is particularly applicable to the study of phase transitions, magnetostriction, and thermal expansion anomalies.

INTRODUCTION

When lecturing to undergraduates, I make it a point to point out that this topic is a tribute to Maxwell, not just as a great physicist, but as a great engineer who had the insight and interest to develop the useful approximations that make calculable capacitors possible, if they are "3-terminal capacitors" with the smaller, defining plate, surrounded by a guard ring.

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One of the most fundamental things we can know about our world is the dependence of energies on the distance between atoms, or, as Faust declares in the opening of Goethe's classic work [1], "what, at the innermost level, holds the world together." Measurements of thermal expansion and magnetostriction are the most direct clues to this dependence in their characterization of the anharmonicity of solids. Capacitance dilatometry is an excellent way to measure these numbers, as well as the changes in interatomic distances at phase transitions. The first extensive work on magnetostriction was that of Becker [2], and an excellent review was provided by Lee [3].

Originally developed by Guy White [4] in Australia, low-temperature capacitance dilatometry was refined by Eric Fawcett while at Bell Telephone Laboratories and then at the University of Toronto. It was used on metal samples that were large enough to form one plate of a capacitor (of area of order 1cm^2) and that could be spark-machined to have flat, parallel surfaces normal to the direction of measurement interest. On moving to a rural campus in Nova Scotia where spark-machining was unavailable, and wishing to study samples that were non-metallic and of odd shapes, the author and his colleagues, Jan Genossar, David Tindall, and Werner Schnepf, developed a dilatometer that met these requirements by having a moving capacitor plate (displaced by the expansion or contraction of a sample in contact with that plate) that was kept parallel to a fixed plate by a pantograph-like arrangement of springs [5]. When extension of this technique to higher temperatures became desirable, the melting of the epoxy and mylar used to insulate the guard ring from the plate became a limiting factor and the weakening of springs at high temperature became an annoyance. This prompted the development of the "tilted plate" capacitance sensor [6]. Refinement of this design using only two materials, a metal and alumina (used for a baseplate and as insulation), has made this a practical tool.

CALCULATING CAPACITANCE

Maxwell [7,8] showed in 1904 that the correction to the expression C=ε_0A/d could be made arbitrarily small if the gap between the guard ring and the smaller plate was sufficiently small, relative to the thickness of the plates.

All students of electricity know that the capacitance of *any* two objects varies as the inverse of the distance between them. The problem is to calculate the capacitance, and in the presence of fringing fields this is a nasty problem. Maxwell showed that the capacitance between an infinite plate and a parallel circular plate of radius, r, and thickness, t, at a spacing, d, with the smaller plate surrounded by a guard ring, with spacing, w, between the plate and the guard ring, is:

$$C = \frac{\varepsilon_0 \pi r^2}{d} + \frac{\varepsilon_0 \pi r w}{d + 0.22w}(1 + \frac{w}{2r})$$

This is true if w is small compared to t, making it possible to calculate the capacitance with a high degree of precision.

The exploitation of the fact that the capacitance of two plates that were tilted relative to one another, i.e. *not parallel*, was just as easy to calculate as that of two parallel plates, by Genossar and Steinitz [5], made the design of capacitance displacement sensors much simpler. Innovative design allowed the elimination of the epoxy insulation that had been used to insulate the guard-ring from the capacitor plate in the designs of White and Fawcett, and the elimination of the springs of the pantograph design, thus enabling the use of these sensors at elevated temperatures. These designs are discussed below.

The capacitance of a capacitor with tilted plates is treated in almost ALL first-year physics textbooks as an irritating problem and an exercise in integration for the students. It is

Michael Steinitz <msteinit@stfx.ca>, Department of Physics, St. Francis Xavier University, P. O. Box 5000, Antigonish, Nova Scotia B2G 2W5

almost always given in the form of a square capacitor, so that the integration can be carried out in strips. This formulation is not only useless, but deceives the students into thinking that this is a horrible problem and that tilt must be avoided at all costs. The answer diverges logarithmically as the gap, d, approaches 0 and most students leave the problem having learned very little.

If, however, the problem is restated with a circular capacitor, the integration becomes difficult, but the physics becomes clear and useful. In this case the student can learn a lot by first examining the silly limits of d=0 and d very large. The interesting point is that as d → 0 the capacitance goes asymptotically to a value which gives the sine of the tilt-angle, a useful number, while at large d the tilt becomes irrelevant. Even more useful is the observation that the capacitance is calculable, analytically, making the capacitor useful as a tool for measuring d. The necessary integral can be found in a table of integrals [9] (Dwight #858.525) or by using Maple or a similar computer mathematics program. If a/r is the sine of the tilt angle, then

$$C = \frac{2\pi\varepsilon_0 r^2 \left(\frac{d^2}{a^2}\right)\left(1-\sqrt{1-\frac{a^2}{d^2}}\right)}{d}$$

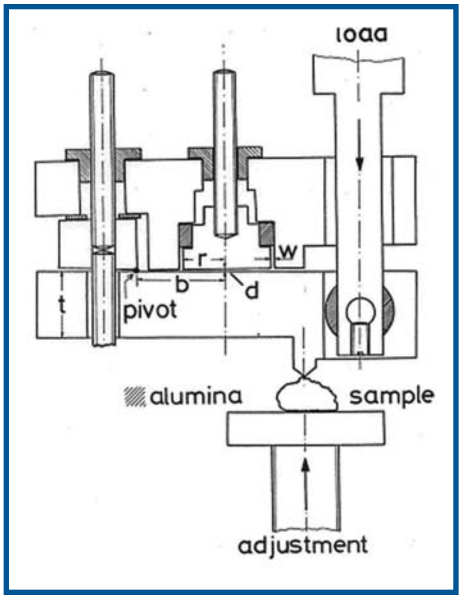

Fig. 1 A drawing showing the construction of a high-temperature dilameter. The pivot point is shown, as well as the point at the top of the drawing where a load can be applied in order to have a uniaxial stress on the sample. Alumina parts are shown in grey, while other parts are metal.

SENSOR DESIGN

Our designs began with an attempt to produce tilted-plate sensors using deposited films of gold on quartz, as indicated in our 1990 paper [6]. The gaps were produced by electron beam lithography and argon ion erosion, because of the need to keep the width of the gap between the plate and the guard ring, w, small relative to the thickness of the plate, t. As the deposited films were only a few thousand angstroms thick, this made a stringent requirement for very small gaps. Although a successful model was produced in cooperation with the National Nano-fabrication Center at Cornell University, our designs subsequently evolved in the direction of massive metal plates insulated by alumina washers easily available commercially (e.g. from Coors Technical Ceramics). Using these ideas, we have been able to keep the guard-ring spacing, w, down to about 0.025 mm with a plate thickness of 3 to 4 mm. As described in our earlier papers [5,6], designs like this allow the construction of a sensor with angstrom resolution, and linearity over a range 10^5 times the resolution.

If one now further refines the design, so that the movable plate pivots about a point a distance b from the center of the movable circular plate, so that b/d is the sine of the tilt angle, then

$$C = \frac{2\pi\varepsilon_0 b^2\left(1-\sqrt{1-\frac{r^2}{b^2}}\right)}{d}$$

Thus C is calculable, proportional, as it should be, to 1/r, with a geometrical factor involving only the radius of the plate, r, and the distance to the pivot point, b. This is the basis for all of our recent designs.

Another key to making this technique a routine measurement has been the availability of commercial capacitance bridges of high resolution. The General Radio 1615 bridge offered 6 decimal places of resolution in the 1960's and is still available today at a cost of about US$30,000. This bridge was balanced by hand for the real and imaginary parts of the impedance and used a lock-in amplifier as a null-detector. The output of the lock-in amplifier could drive a chart-recorder and could later be coupled to a computer. A major improvement came with the availability in the 1980's of the Andeen-Hagerling capacitance bridge, which offered 8 decimal places of resolution, was self-balancing, and communicated directly with a computer via the IEEE-488 interface bus at a cost of around US$10,000.

Our most recent design allows the application of a uniaxial stress along the measurement direction, as shown in Figure 1.

This design incorporates the lessons learned over many years of use of these dilatometers.

Figure 2 shows the small plate, above, with the gap to the guard ring clearly visible as the smaller circle, with the large moveable plate below.

Figure 3 shows a high temperature capacitance dilatometer mounted on a differential micrometer for cali-

Fig. 2 Photograph of the sensor parts of the dilatometer of Figure 1. The upper piece contains the fixed circular plate, separated from the guard ring by the gap, which can be seen as the smaller circle. The lower piece is the moveable plate.

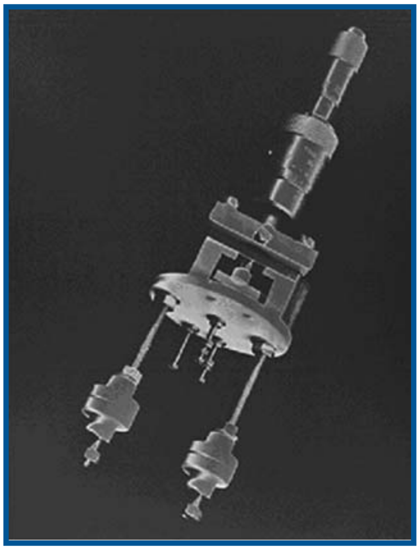

Fig. 3 Photograph of an older version of high-temperature dilatometer, shown attached to a differential micrometer for calibration. The small screws seen protruding from the alumina plate are for attachment of electrical leads, and the large screws are for mounting. In use, this dilatometer is inverted, i.e. hangs down from the alumina plate shown in the photograph.

bration purposes. The dilatometer has resolution better than 1 Angstrom (10^{-10} m), the differential micrometer has a resolution of .0005 mm ($5x10^{-7}$ m), while the coarse adjustment of the micrometer has a resolution of 0.01 mm (10^{-5} m). The dilatometer itself is linear over a range of 0.3 mm, i.e. over five orders of magnitude of its resolution. It offers measurements of changes of sample length with Angstrom resolution at temperatures up to 1000 degrees Celsius, with a small compact sensor, located at the sample position and at the sample temperature.

SENSOR MATERIALS

We have learned a great deal about materials in the course of the development of these dilatometers. Working in vacuum at high temperatures, we have had success with stainless-steel and titanium sensors and alumina washers and baseplates, but these materials are limited to temperatures below their individual structural phase transitions. We thought that Molybdenum would be an ideal material, having a very high melting point and no phase transitions. We therefore developed machining techniques for Mo, a capability found very rarely in the world, only to find that the sublimation of surface layers of molybdenum oxide resulted in the deposition of fine layers of Mo on all the alumina surfaces, short circuiting the capacitance leads, the insulating washers, and generally making a terrible mess! Thus the metal chosen for the sensor must be evaluated for the specific temperature range of interest.

OTHER SENSORS

References to other capacitance dilatometers are included in references 5 and 6 and other dilatometers are commercially available, usually relying on linear variable differential transformers (LVDT's) and push-rods to transmit the motion to a sensor outside the sample environment. An LVDT-based dilatometer allowing the application of stress was described by Cherepin *et al.* [10]. An elegant dilatometer based on the tilted-plate design was reported by Rotter *et al.* [11] in 1998.

A FEW INTERESTING RESULTS

We have made many interesting studies using these instruments, including industrial reports on measurements of annealing in radiation-damaged diamonds and swelling of adhesives. Among the most satisfying were the observation of fluctuations in periodicity in holmium [12] by measuring the "noise" in the length changes and its disappearance in "locked-in" states, and recent measurements on the spin-reorientation transition in cobalt [13] and on the order-disorder transition in sodium nitrate [14].

ACKNOWLEDGEMENTS

I am very grateful for the assistance in all this work of my colleague, designer and machinist, Werner Schnepf, my colleagues, Jan Genossar and David Tindall, and my many students who have slaved over short-circuited sensors, malfunctioning electronics, and other challenges to produce excellent data. Recent students have included Mark Lumsden, Michael Dunlavy, Daniel Beaton, Michael Lewis, Nicole MacDonald and Patrick Clancy.

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