CAPACITANCE DILATOMETRY MOVES FROM CRYOGENIC TO HIGH TEMPERATURES

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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.

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, "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, and an excellent review was provided by Lee. Originally developed by Guy White 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 1cm²) 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 Schrepp, 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. 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. 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 showed in 1904 that the correction to the expression \( C = \varepsilon_0 \frac{A}{d} \) 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:

\[
C = \frac{\varepsilon_0 \pi r^2}{d} + \frac{\varepsilon_0 \pi rw}{d + 0.22w} \left( 1 + \frac{w}{2r} \right)
\]

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, 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
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 (Dwight #858.525) or by using Maple or a similar computer mathematics program. If a/r is the sine of the tilt angle, then

\[
\frac{2\pi \varepsilon_0 r^2}{d} \left( 1 - \frac{a^2}{d^2} \right)
\]

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 designs began with an attempt to produce tilted-plate sensors using deposited films of gold on quartz, as indicated in our 1990 paper. 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 massless 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, designs like this allow the construction of a sensor with angstrom resolution, and linearity over a range 10^5 times the resolution.
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].

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REFERENCES

1. J.W. v. Goethe, Faust: Eine Tragödie, (Nacht "... Daß ich erkenne, was die Welt Im Innersten zusammenhält"), (Weimar Tübingen: Cotta, 1808).