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Electrostriction Measurements on Low Permittivity Dielectric Materials

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Abstract

To measure the electrostrictive effects in low permittivity materials, extremely sensitive instrumentation is required. A modified compressometer for resolving fractional changes in capacitance on the order of 10^{-6} *is used in this work, along with a modified single beam interferometer capable of subangstrom resolution in displacement. For the compressometric method, a high sensitivity capacitance bridge, GenRad 1615, is coupled with two lock-in amplifiers to detect attofarad (*10^{-18}*F) level capacitance changes caused by in-phase cyclic uniaxial stresses on samples. The interferometer is a Michelson –Morley type instrument modified to detect changes in interference fringe intensity for very small changes in path length. The measurements confirmed by both techniques are used to establish a set of reliable and accurate data of electrostriction coefficients for low permittivity materials. Using these recent data, along with widely accepted data on ferroelectric materials and soft polymers, the linear relationship between electrostriction coefficient* (Q) *and the ratio of elastic compliance over dielectric permittivity* ($s/\varepsilon_0\varepsilon_r$) *is obtained. This leads to an effective way to predict the electrostriction coefficient in dielectric materials. © 1999 Elsevier Science Limited. All rights reserved*

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1 Introduction

Electrostriction is the fundamental mechanism of electromechanical coupling in all insulator materials. Its magnitudes can range from very minute in low permittivity dielectrics to very large in ferroelectrics. Theoretical studies of this fundamental phenomenon are handicapped by the absence of reliable and accurate experimental data in simple low permittivity dielectrics. In practical applications, electrostrictive stresses can cause breakdown in insulator materials in microelectronics and high voltage devices.[1] Therefore, this study was undertaken with the primary goal of establishing reliable and accurate electrostriction coefficients for low permittivity dielectrics.

Electrostriction is defined as the quadratic coupling between strain (x) and electric field (E), or between strain and polarization (P). This is a fourth-rank tensor expressed by the following relationships:[2,3]

$$x_{ij} = s^{E}_{ijkl}X_{kl} + M_{ijmn}E_mE_n \tag{1}$$

$$x_{ij} = s^{P}_{ijkl}X_{kl} + Q_{ijmn}P_mP_n \tag{2}$$

where s_{ijkl} is the elastic compliance tensor under appropriate boundary conditions and X_{kl} is the elastic stress component. From these, the 'direct' effect Q and M electrostriction coefficients are defined as:

$$M_{ijmn} = 1/2(\mathrm{d}^2x_{ij}/\mathrm{d}E_m\mathrm{d}E_n)_X \tag{3}$$

$$Q_{ijmn} = 1/2(\mathrm{d}^2x_{ij}/\mathrm{d}P_m\mathrm{d}P_n)_X \tag{4}$$

Alternatively, by application of Maxwell relations to the above equations, one can derive the 'first converse' effect Q and M coefficients in terms of the dielectric susceptibility (χ_{ij}) and its inverse, the dielectric stiffness tensor (η_{ij}) as:

$$M_{ijmn} = 1/2(\mathrm{d}\eta_{ij}/dX_{mn})_P \tag{5}$$

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$$Q_{ijmn} = -1/2(\mathrm{d}\chi_{ij}/\mathrm{d}X_{mn})_P \quad (6)$$

The ‘second converse’ effect is the polarization dependence of the piezoelectric voltage coefficient (g_{nij}) and can be defined as:

$$Q_{ijmn} = \delta g_{nij}/\delta P_m \quad (7)$$

$$M_{ijmn} = \varepsilon_0(\varepsilon_{ij} - 1)^2 \delta g_{nij}/\delta P_m \quad (8)$$

The direct and converse electrostriction effects are of importance because they offer two independent and equivalent techniques for electrostriction measurements. Measurements confirmed by both the direct and converse methods would result in more reliable and accurate electrostriction coefficients in materials of interest, such as low permittivity dielectrics.[1,3,4]

2 Electrostriction Measurement Techniques

Various techniques can be used to determine electrostriction coefficients. Widely used experimental techniques include the strain gauge method, the capacitance dilatometer, and laser ultradilatometer based on Michelson interferometer. In this work, two independent experimental approaches were used to determine electrostriction coefficients: a modified Michelson–Morley interferometer, measuring the strains induced in materials in response to applied fields or induced polarizations using the direct effect, and a dynamic compressometer, measuring the change in permittivity under appropriate stress using converse effect.

2.1 The dynamic compressometer

In the converse method the change in permittivity was measured as a function of stress using a mechanically stable compressometer isolated from electrical and mechanical noises. The system was designed to apply a homogeneous pressure on the sample surface from a completely uniaxial loading. The sample was held between Plexiglas ram extenders. This prevented shear deformations arising from elastic mismatch between the metal ram and samples.

Significant modifications were made to the original compressometer design[1] involving a cyclic loading system and a revised measurement technique. The redesigned compressometer is shown in Fig. 1. A stepping motor and spring were used to apply cyclic stress, which was uniaxially transferred to the sample through extension rams. The capacitance of the sample was first measured to a resolution of 0·0001 pF on a GenRad 1615 capacitance bridge. A Stanford SR830 DSP Lock-In Amplifier was used to read the bridge balancing null. The output from this amplifier was then used as input for a second lock-in amplifier, which was electronically locked to the revolving frequency of the stepping motor using a reflecting sensor. During cyclic loading, the first lock-in amplifier produced an electrical signal proportional to capacitance change, then a precise fractional change in capacitance of the sample was calculated from the output of the second lock-in amplifier. Thermal and mechanical noises were electronically rejected from the measurements with use of both lock-in amplifiers.

These compressometric measurements were performed at 1 kHz, corresponding to the frequency of the first lock-in amplifier. To optimize the measurement conditions, a stepping motor frequency was appropriately set to provide a cyclic loading at 0·4 Hz, at which a second lock-in amplifier was also automatically set through a sensor. Throughout the experiment, a maximum load of 9·96 kg was used to achieve relatively high level of capacitance changes.

2.2 The interferometric ultradilatometer

A modified Michelson–Morley interferometer was constructed specifically for this study.[5] In the interferometric method, the sample under investigation was subjected to an AC field, and was interrogated with a polarized beam from a Helium–Neon laser. The path difference between the beam reflected from the sample surface and a reference beam produced interference, and resultant intensity variations were sensed by a photodiode. A schematic of the system is shown in Fig. 2. It is a compact system with very light and rigid optical components. The total optical path length in the system is small, which is of utmost importance in reducing mechanical noise. The difference in the lengths of the optical path traveled by the two interferometer arms was kept small by mounting the sample, laser, photodetector and the electrostrictive stack on the circumference of a Plexiglas ring ~0·5 m in diameter. This minimized laser coherence length problems, reduced thermal drift, and compensated for beam divergence problems. The system is mounted on a pneumatically supported table to reduce the effect of ambient vibrations. Temperature and electrical stability was achieved by fitting the sample with an aluminum lined foamboard lid. The laser used is a 2 mW polarized He–Ne laser (Uniphase, $\lambda = 632{\cdot}8$ nm). The reference mirror, that reflects the reference beam, is mounted on an electrostrictive actuator stack connected to a feedback loop. This loop stabilizes the system at a point where the path difference is $\lambda/4$, the ‘π/2 point’,

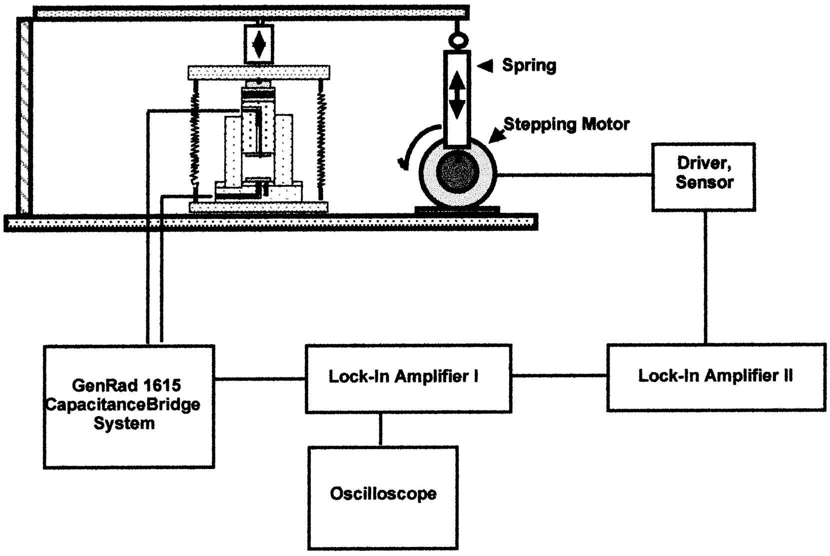

Fig. 1. Schematic of dynamic compressometer.

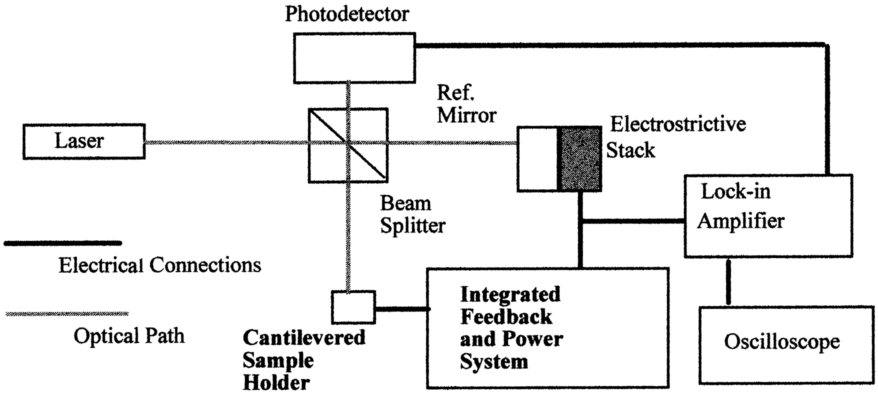

Fig. 2. Schematic of laser interferometer.

where light intensity change is maximized for a small change in the displacement Δd of the sample surface. When an AC field is applied to the sample, small sinusoidal displacements are obtained, giving interference intensity changes at the point of detection. This optical signal is converted to a voltage change by a biased photodiode (Motorola MRD500). The photocurrent is then converted to a voltage using a high frequency current to voltage converter with a gain of $10^4\,\mathrm{VA}^{-1}$. This voltage is detected by a lock-in amplifier (Stanford Research Systems 830) as V_{out}, an rms value. The interference fringe shifts are observed using an oscilloscope (Hewlett–Packard 54600B).

3 Experimental Procedure

3.1 Compressometry euations

The equation for the capacitance of a parallel plate insulator, $C = \varepsilon_0 \varepsilon_r A/d$, may be separated and differentiated with respect to stress applied to give:[1]

$$\frac{\delta \ln \varepsilon_r}{\delta X} = \left[\frac{\Delta C/C}{\Delta X} - \frac{\delta \ln(A/D)}{\delta X}\right] = B_i(\text{measured}) - S_i \tag{9}$$

where S_i are linear combinations of elastic compliances, and B_i pressure dependencies of the capacitance for the given cut. M' and Q', the coefficients for the given cut for the first converse effect, are given by:

$$Q = [\varepsilon_r / 2\varepsilon_0(\varepsilon_r - 1)^2][B_i - S_i] \tag{10}$$

$$M = (\varepsilon_0 \varepsilon_r / 2)[B_i - S_i] \tag{11}$$

3.2 Interferometry equations

For monochromatic light with a wavelength λ, interfering with a reference beam, the interference intensity detected may be expressed as:[3]

$$I = Ip + Ir + 2IpIr\sin(4\pi\Delta d/\lambda) \qquad (12)$$

where Ip and Ir are the intensities of the probe and reference beams, respectively. For small displacements Δd about the $\pi/2$ point, one may rewrite the intensity detected as:

$$I = 1/2(I_{\max} + I_{\min}) + 1/2(I_{\max} - I_{\min})\sin(4\pi\Delta d/\lambda) \qquad (13)$$

For such a small displacement, one may assume sin $x \sim x$. This implies the interference intensity change is linearly proportional to the induced displacement. The displacement may now be expressed as:

$$\Delta d = \left(\frac{\lambda}{\sqrt{2}\pi}\right)\left(\frac{V_{\text{out}}}{V_{p-p}}\right) \qquad (14)$$

This displacement is plotted as a function of applied voltage to calculate the electrostriction coefficients. For this direct method, the attractive coulombic forces between the free charges on the electrodes of the sample causes a change in sample dimension. This is called 'Maxwell strain'. For a parallel plate capacitor, the attractive stress is given as:[3]

$$X_m = -1/2(\varepsilon_{11}\varepsilon_0 E^2) \qquad (15)$$

A corresponding value to the measured strain is added to correct for the Maxwell strain. Hence, the true direct effect electrostrictive coefficients are given by:

$$M'_{11} = M'_{11}(\text{measured}) + 1/2(s'_{11}\varepsilon_0\varepsilon_{11}) \qquad (16)$$

3.3 Sample preparation

Single crystals of orientations, <100>, <110>, <111>; were used for cubic samples CaF_2, BaF_2, SrF_2, and $KMnF_3$. For MgO, BeO, and $CaCO_3$, single crystals with <100> orientation were obtained.

Preliminary compressometric studies were also carried out on ceramics, such as ZrO_2, Al_2O_3, AlN, and Si_3N_4 and on polymers, such as polystyrene (PS) and polyethylene terephthalate (PET).

For the compressometer, the samples are thin discs with plane parallel faces. The sample dimensions are approximately 20 mm in diameter and 1 mm in thickness. On the other hand, 1 mm^3 cubes are used in the interferometer. Gold electrodes are sputtered on samples polished to optical finish.

4 Experimental Results

Q coefficients for the samples from both techniques are given in Table 1. It may be seen in the tabulation that all results agree well. This helps us achieve our primary aim of establishing the reliable and accurate electrostriction coefficients, particularly in low permittivity dielectrics.

5 Prediction of Electrostriction Coefficients

Under the action of an applied electric field, the cations and anions in a crystal structure are displaced in opposite directions by an amount Δr. This displacement is responsible for the electric polarization, the dielectric permittivity, and the electrostrictive strain. We may express the relation between these as:

$$Q = x/P^2 \sim (\Delta r)/(\Delta r)^2 = 1/\Delta r \sim 1/\varepsilon_0\varepsilon_r \qquad (17)$$

Large strains in compliant solids such as polymers can introduce large changes in the dielectric stiffness and in anharmonic potentials. Electrostrictive strain is displacive and acts against the elastic forces in a material. Introducing the elastic compliance s into the Q versus $1/\varepsilon_0\varepsilon_r$ relationship can better express the correlation between these properties. Through the first converse effect, electrostriction is proportional to the change of dielectric stiffness η with stress X, which may be expressed as:

$$Q \sim (\Delta\eta)/(X) \sim x/(x/s) \sim s \qquad (18)$$

Table 1. Results from direct and converse effects[4]

Material		*Direct* Q m^4/C^2	*Converse* Q m^4/C^2
BaF_2	Q_{11}	−0·33	−0·31
	Q_{12}	−0·29	−0·29
	Q_{44}	1·46	1·48
CaF_2	Q_{11}	−0·48	−0·49
	Q_{12}	−0·48	−0·48
	Q_{44}	−0·33	−0·33
SrF_2	Q_{11}	−0·33	−0·33
	Q_{12}	0·38	0·39
	Q_{44}	2·01	1·90
$KMnF_3$	Q_{11}	0·51	0·49
	Q_{12}	−0·09	−0·10
	Q_{44}	1·16	1·15
MgO	Q_{11}	0·33	0·34
BeO	Q_{11}	1·45	1·48
Calcite	Q_{11}	1·20	1·19
Al_2O_3	Q_{33}	–	−0·20
ZrO_2	Q_{33}	–	0·06
AlN	Q_{33}	–	−0·27
Si_3N_4	Q_{33}	–	−0·36
PS	Q_{33}	–	−35
PET	Q_{33}	–	−22

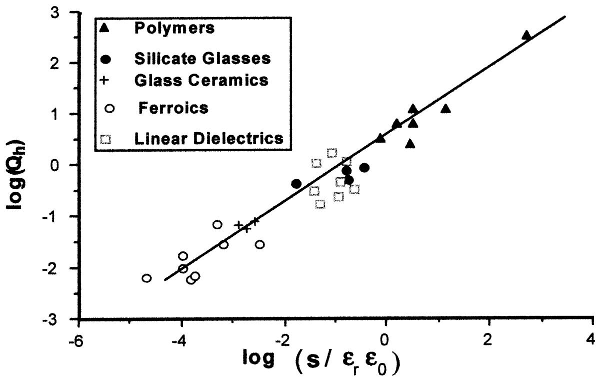

Fig. 3. Variation of log Q_h(m^4/C^2)with log ($s/\varepsilon_0\varepsilon_r$).

Empirically, log (Q_h) is found to vary linearly with log ($s/\varepsilon_0\varepsilon_r$) (Fig. 3). This combination of the compliance and the dielectric properties of materials may be used to predict the magnitude of electrostriction in insulators.

6 Conclusions

In this present work, two extremely sensitive instrumentations, a modified single beam interferometer and a dynamic compressometer, were described. The measurements confirmed by both techniques were used to establish a set of reliable and accurate data of electrostriction coefficients for low permittivity materials. Finally, the linear relationship between electrostriction coefficient (Q) and the ratio of elastic compliance over dielectric permittivity ($s/\varepsilon_0\varepsilon_r$) was obtained. This leads to an effective way to predict the electrostriction coefficient in dielectric materials.

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