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Non Linear Behavior of the Permittivity and of the Piezoelectric Strain Constant Under High Electric Field Drive

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Abstract. Two methods are proposed and compared for the measurement of the converse piezoelectric strain constant: A Laser Doppler Vibrometer Technique and a Capacitance Probe. The complete results obtained with the Capacitance Probe device are given for ''hard'' and ''soft'' PZT samples between 1 Hz – 1 kHz and up to 700 Vp with and without a dc offset bias. The permittivity is also studied under various conditions of electric drive (ac, dc, ac + dc . . .) with the help of a LCR meter and a Schering Bridge. Many analogies between the dielectric non linearities and the piezoelectric one's are observed. In both cases the high field behavior follows the empirical Rayleigh law providing that the dc and ac influences would be separated.

Keywords: ferroelectric ceramics, non-linearities, high field, piezoelectric strain constant

1. Introduction

The most often used coefficient for characterizing the piezoelectric response of a ceramic is the piezoelectric charge constant d_{33}. This coefficient is easily measured using the direct piezoelectric effect: for sufficiently low levels of dynamic mechanical stress and at frequencies far below the resonance frequency of the test specimen ($f \leq 1000\,\mathrm{Hz}$), and at zero electric field the following relationship is valid: $D_3 = d_{33}T_3$, where D_3 is the electrical induction ($\mathrm{C\ m^{-2}}$) (e.g. such an arrangement is found in Berlincourt Piezometer).

The d_{33} value obtained by this way is very close to the d_{33} coefficient determined from electromechnical longitudinal coupling coefficient, permittivity and elastic constant (IEEE Standard Procedure [1]). However such a low level coefficient cannot be used for describing the charge release in piezoceramics driven at high mechanical stress or the strain of piezoceramics driven at high ac electric field. Results have been given on the d_{33} behavior under high static mechanical stress [2,3] and high static electrical field [4,5] but there are few results on the dynamic behavior of d_{33} under high ac electrical field [6].

D. V. Taylor and D. A. Hall have extensively studied the ac high field non linearity of the permittivity [7–10] and D. Damjanovic et al. have studied the high stress non linearity of d_{33} under ac and dc conditions [11–13]. Both found a Rayleigh behavior of the coefficients.

The purpose of this study is to measure the converse piezoelectric coefficient d_{33c} at various electric drive fields E_3 and frequencies (d_{33c} is defined as the ratio $S_3/E_3 = u_3/V_3$ where S_3 is the relative displacement, u_3 the displacement and V_3 the electric voltage). Results obtained on ''hard'' and ''soft'' PZT samples are compared with permittivity measurements.

It appears that the strain and piezoelectric non linearities as a function of the dc and ac electric fields are very similar and follow the Rayleigh law.

2. Experimental Procedures

A direct measurement of very small displacements is difficult because of the very low signal to noise ratio. However, the use of a high performance lock-in amplifier allows such measurements.

A Laser Doppler Vibrometer Technique (LDVT) measurement device is given in Fig. 1. The vibration velocity measurement unit is a Polytec Vibrometer composed of an optic head OFV 302 and a control unit OFV 3000. A DSP lock-in amplifier Stanford SR 830 associated with the Polytec measurement unit, measures the magnitude and phase of the fundamental and harmonics of the vibration velocity.

A non contact measurement of the displacement is also achieved with the help of a capacitive comparator FOGALE MC 940. The sensor must be facing a ground electrode; so a non-differential measurement with a single sensor has been chosen. As with the LDVT method an accurate measurement will require a zero displacement of the rear electrode subjected to the high voltage.

The bandwidth is dc to 10 kHz but the measurements have been restricted to 1 Hz − 1 kHz.

A set of poled disk shaped PZT ceramics ($\phi = 10\,\text{mm}, l = 1\,\text{mm}$) was obtained from Morgan Matroc (Wrexham U.K.). The soft type was PZT 5A and the hard type was PZT 4D.

In order to obtain a good damping of its rear face, while allowing a free lateral strain the disk is laid on a plastic support and a possible film of air or detachment between the plate and the support is eliminated by using a very viscous grease (Dow Corning high vacuum grease) or a double face gummed ribbon (Fig. 2).

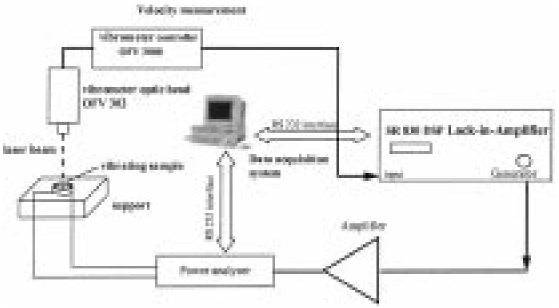

Fig. 1. LDVT measurement device.

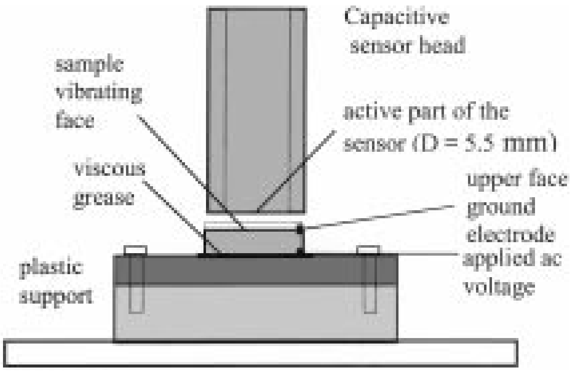

Fig. 2. Piezoelectric ceramic holding.

With this sample holding the reproducibility of the measurements is quite good.

Results given in Fig. 3 show that the difference between the LDVT and Capacitive devices is around 4% in the frequency range 100 Hz–1000 Hz.

As a conclusion, the two methods don't require the use of an antivibration table and present a good accuracy. However the capacitive device is chosen for the next measurements because it allows driving at very low frequency

The high level permittivity and loss tangent are measured with and without a dc bias with the help of a LCR Meter HP 4284A using very thin samples (0.3 mm in thickness, 6.35 mm in diameter). For other samples a Schering Bridge is used for ac measurements at 1 kHz without a dc bias.

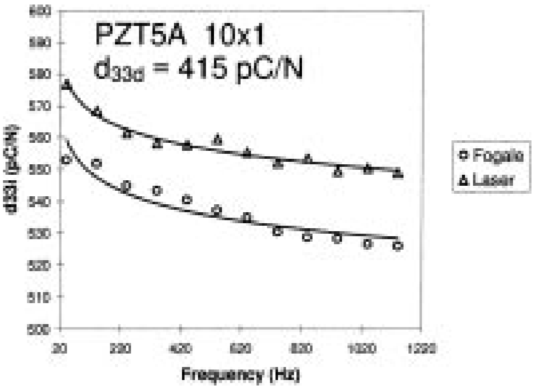

Fig. 3. LDVT and CAPACITIVE SENSOR measurements: d_{33c} versus frequency for $E = 200\,\text{V mm}^{-1}$ (rms).

3. Results

3.1. *Measurement of the Converse Piezoelectric Coefficient d_{33c}*

3.1.1. Influence of the ac Electric Field and the Frequency. Measurements of d_{33c} on samples 1 mm in thickness are given in Figs. 4 and 5 as a function of the rms electric field and of the frequency. The ac driving voltage is sinusoidal without a dc offset voltage.

For the ''soft'' type composition a logarithmic frequency dependence of d_{33c} is observed but d_{33c} is noticeably increasing as a function of the driving electric field. For the ''hard'' type composition d_{33c} is nearly constant as a function of frequency whereas the variations as a function of the applied field are much smaller than in soft one. Results summed up in Table 1 show that the value of d_{33c} extrapolated with the experimental results at $E = 0$ is close to the direct piezoelectric coefficient (d_{33} Berlincourt) measured at the same frequency. The harmonic distorsion ratio of the displacement signal increases with the applied field (both even and odd harmonics are observed).

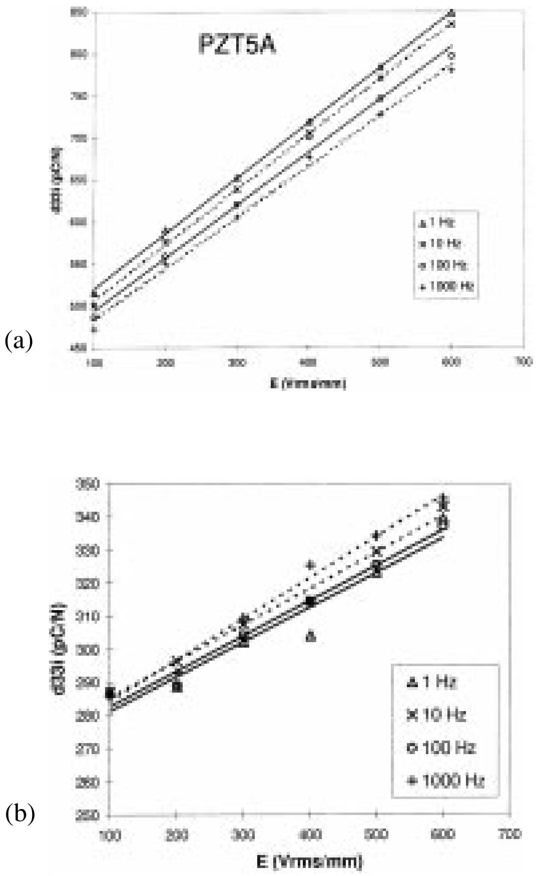

Fig. 4. d_{33c} versus the applied electric field at various frequencies: a) PZT5A, b) PZT4D.

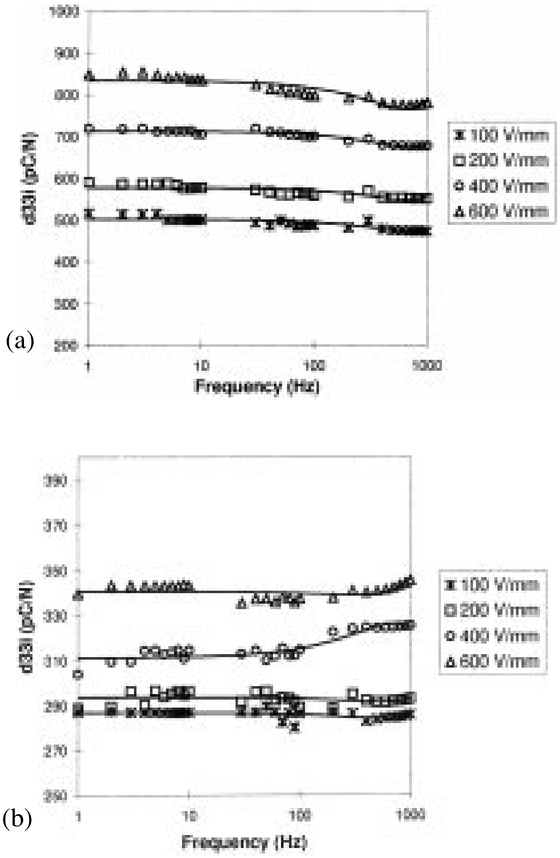

Fig. 5. d_{33c} versus frequency for various electric fields: a) PZT5A, b) PZT4D.

3.1.2. Influence of a dc Bias. Figure 6 shows the variation of d_{33c} when the ceramics are subjected both to an ac electric field and a superimposed offset bias $V_{dc} = \pm V_{peak}$ such as only the positive region (full line) or the negative region (dotted line) is covered : In every case the variations of d_{33c} versus the applied ac field are more important with the negative offset.

Table 1. Frequency and electric field dependence of d_{33c} for soft and hard PZT (E_p = electric field peak value)

Sample	d_{33} Berlincourt	d_{33c} 120 Hz extrapolated	$\Delta d_{33c}/d_{33c}$ (%) between 1 Hz—1 kHz		$\gamma_{ac} = \frac{1}{d_{33c}}\frac{\delta d_{33c}}{\delta E_p}$ 10^{-7} mV^{-1}		d_{33c} Harmonic distorsion ratio (%) 120 Hz	
$\phi = 10$ mm	120 Hz	at $E = 0$	100	600			100	400
$t = 1$ mm	pCN^{-1}	pCN^{-1}	V/mm	V/mm	1 Hz	1 kHz	V/mm	V/mm
PZT 5A	417	423	− 8	− 8	10.2	9.8	1.4	2.6
PZT 4D	312	283	# 0	+ 0.7	2.7	2.9	0.6	2.4

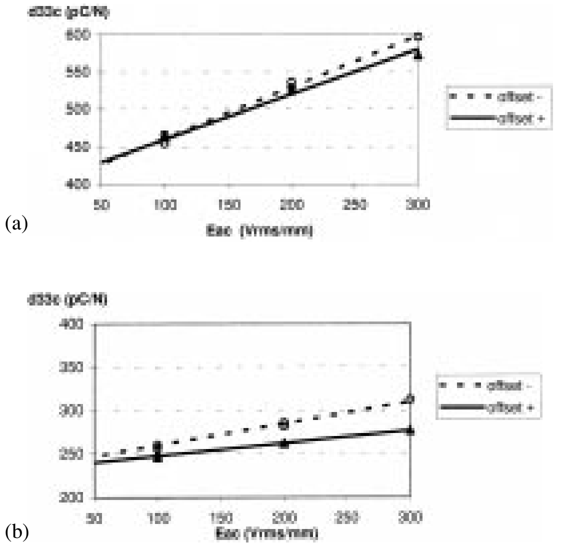

Fig. 6. d_{33c} versus the ac field with positive (–) and negative (- - -) offset ($f = 1$ kHz, $E_{dc} = \pm E_p$): a) PZT5A, b) PZT4D.

At constant ac drive and in function of the dc offset, d_{33c} is slightly decreasing except for PZT 4D at positive offset (Fig. 7).

3.2. *Permittivity Measurements*

3.2.1. Influence of an ac Field. As shown in Fig. 8 the increase of the permittivity as a function of the ac field is similar to that observed with d_{33c}.

3.2.2. Influence of an dc Bias. The influences of a superimposed dc offset ($E_{dc} = \pm E_p$) (Fig. 9) and of a variable dc offset at constant E_{ac} (Fig. 10) are also similar to those observed with d_{33c}.

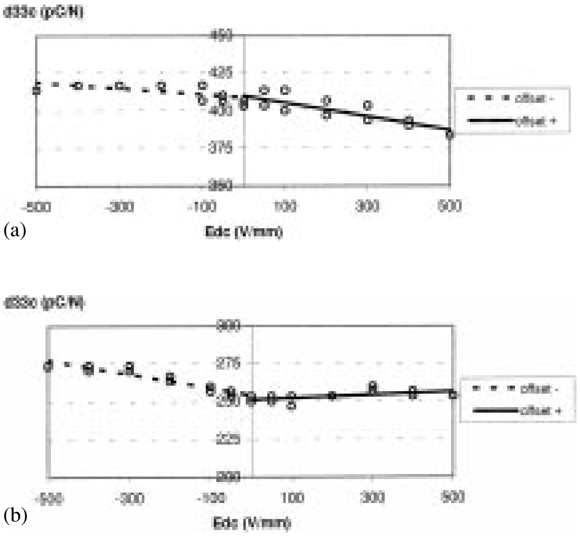

Fig. 7. d_{33c} versus the dc field ($f = 1$ kHz, $E_{ac} = 30$ V/mm): a) PZT5A, b) PZT4D.

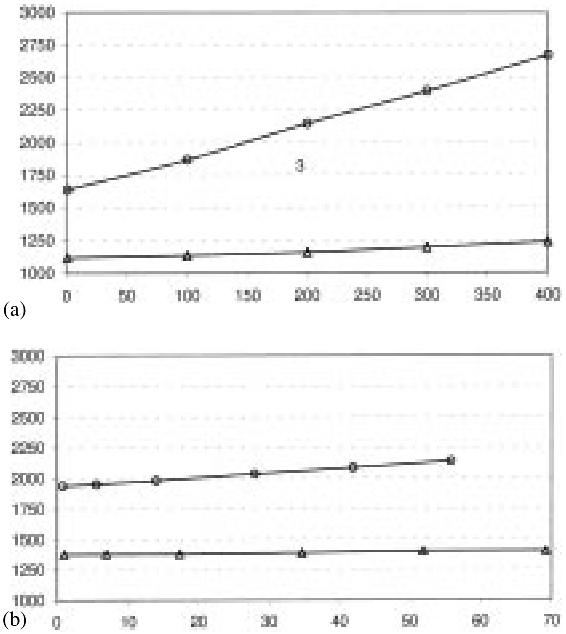

Fig. 8. ε_{33r} versus applied electric field ($f = 1$ kHz): a) Schering bridge (samples 1 mm in thickness), b) Impedancemeter (samples 0.3 mm in thickness) ○ PZT5A and △ PZT4D.

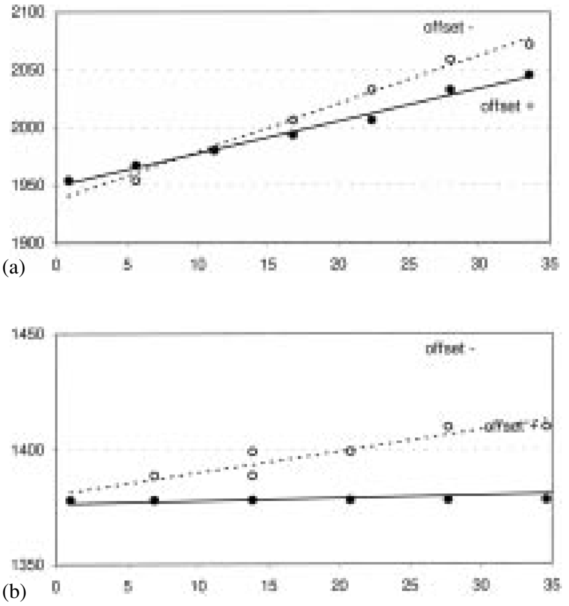

Fig. 9. ε_{33r} versus the ac field with positive (–) and negative (- - -) offsets ($f = 1$ kHz, $E_{dc} = \pm E_p$): a) PZT5A, b) PZT4D.

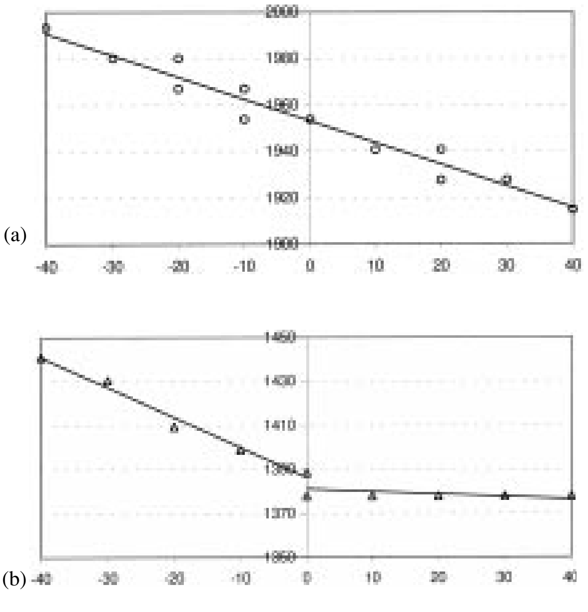

Fig. 10. ε_{33r} versus the dc bias ($f = 1$ kHz, $E_{ac} = 3$ V/mm): a) PZT5A, b) PZT4D.

4. Phenomenological Study of the Non Linear Behavior of the Piezoelectric Strain Constant and of the Permittivity Under High Electric Field

D.V. Taylor and D. Damjanovic have shown that a linear ε^T_{33} (E_p) relationship was found, which is analogous to the Rayleigh law in ferromagnetic materials [7]. Such a non linearity would be due to the movement of 90° domain walls across an array of pinning effects (acceptor, oxygen vacancy . . .).

Our experimental results show that d_{33c} and ε_{33c} follow also a Rayleigh law as a function of the electric field but the effects of a static dc field and of a dynamic ac field have to be separated.

So the variation of d_{33c} can be written as follows : $d_{33c} = d_{33oc}[1 + \gamma_{dc}E_{dc} + \gamma_{ac}E_p]$ where E_{dc} is the static electric field and E_p the peak value of the ac field.

In a similar way the variation of the permittivity can be written as :

$$\varepsilon^T_{33} = \varepsilon^T_{33o}\left[1 + \alpha_{dc}E_{dc} + \alpha_{ac}E_p\right]$$

- $$\gamma_{ac} = \frac{1}{d_{33oc}}\frac{\delta d_{33c}}{\delta E_p} \text{ and } \alpha_{ac} = \frac{1}{\varepsilon^T_{33o}}\frac{\delta \varepsilon^T_{33}}{\delta E_p}$$

  are calculated from studies at zero or constant offset E_{dc}

- $$\gamma_{dc} = \frac{1}{d_{33oc}}\frac{\delta d_{33c}}{\delta E_{dc}} \text{ and } \alpha_{dc} = \frac{1}{\varepsilon^T_{33o}}\frac{\delta \varepsilon^T_{33}}{\delta E_{dc}}$$

  are calculated from studies at constant ac electric field : γ^+_{dc}, α^+_{dc} and γ^-_{dc}, α^-_{dc} are respectively the coefficients for a positive or a negative offset bias
- Studies with a positive offset voltage $E_{dc} = E_p$ allow to calculate the coefficients γ^+_{off} and α^+_{off} (and with a negative offset voltage $E_{dc} = -E_p, \gamma_{off}{}^-$ and $\alpha_{off}{}^+$) whose theoretical values are :

$$\gamma^+_{off} = \gamma_{ac} + \gamma^+_{dc}, \alpha^+_{off} = \alpha_{ac} + \alpha^+_{dc},$$
$$\gamma^-_{off} = \gamma_{ac} - \gamma^-_{dc}, \alpha^-_{off} = \alpha_{ac} - \alpha^-_{dc}.$$

The experimental values of the γ and α coefficients are given in Table 2 as well as the experimental and theoretical values of the coefficients γ_{off} and α_{off} which are in good agreement.

The influences of the ac or dc electric field on d_{33} and ε^T_{33} are the same. At last the voltage constants

Table 2. Values of coefficients γ and α

Coefficient 10^{-7} m/V	γ_{ac}	γ_{dc}^{+}	γ_{dc}^{-}	γ_{off}^{+} theo	γ_{off}^{+}	γ_{off}^{-} theo	γ_{off}^{-}
PZT 5A	9.8	− 1	− 0.6	8.8	8.4	10.4	10.5
PZT 4D	2.9	+ 1.2	− 2.3	4.1	4.4	5.2	7.4
Coefficient 10^{-7} m/V	α_{ac}	α_{dc}^{+}	α_{dc}	α_{off}^{+} theo	α_{off}^{+}	α_{off}^{-} theo	α_{off}^{-}
PZT 5A	13*	− 1.8	− 1.8	11.2	10.2	14.8	15.2
PZT 4D	1.6**	0	− 2.7	1.6	1.5	4.3	4.6

*10.6 with Schering Bridge.
**1.3 with Schering Bridge.

$g_{33} = d_{33}/\varepsilon_{33}^{T}$ will be nearly independent of the electric field.

At low field there is no evidence of a threshold field and at high field we have not approached the coercive field level.

5. Conclusion

As a conclusion both the LDVT and Capacitive Sensor methods coupled with a DSP lock in amplifier have proved to be highly suitable for dynamic field induced strain measurements. Linear d_{33c} (E) and ε_{33}^{T} (E) relationships were obtained for both soft and hard PZT, giving a clearly Rayleigh behavior. However the effects of a dc or ac field have to be differentiated and specific Rayleigh coefficients are calculated in each case.

Similar behavior of d_{33c} (E) and ε_{33}^{T} (E) suggests that they are controled by the same 90° domain wall translation mechanism.

Acknowledgment

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