ELECTROMECHANICAL NONLINEARITY OF FERROELECTRIC CERAMICS AND 90^0 DOMAIN WALL MOTIONS

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ABSTRACT

By using an optical interferometer and other more conventional methods, the responses of piezoelectric and dielectric coefficients of PZT ceramics to a.c. electric fields have been directly investigated. The experimental results demonstrate the importance of electromechanical non-linearities present in the ferroelectric ceramics.

A phenomenological theory, based on a model of Arlt et al., has been extended to describe the extrinsic contribution to piezoelectric, elastic and dielectric coefficients. This can be attributed to both the linear and nonlinear vibrations of 90^0 domain walls in tetragonally distorted ferroelectric ceramics. The proposed theory shows qualitative agreement with the experimental results

I. INTRODUCTION

Poled ferroelectric ceramics are widely used as transducers, resonators, actuators, motors, and sonars. One of their most fundamental limitations for practical use are the mechanical nonlinear effects, which occur at high drive levels. The strong nonlinear behavior in ferroelectric materials is due in large part to the effect of the mechanical stress and electric field on the configurations of the domains in the ceramics[1,2].

But very little work has been done to describe the behavior of electromechanical nonlinearity in terms of domain wall motion, although the domain wall motion plays a predominant role at certain frequencies span in ferroelectric ceramics.

In fact, studies[3,5] on PZT ceramics have shown that 60% - 70% of the values of piezoelectric, dielectric, and elastic coefficients come from the extrinsic contributions which are produced by movements of non-180° domain walls and interphase boundaries.

Therefore, from the technological point of view, it is necessary to study the relationship between electromechanical properties and domain structures in ferroelectric ceramics in order to optimize the choice of piezoceramics suitable for transducers, actuators, resonators and acoustic wave devices.

In the present work, the relationships between domain wall motions and piezoelectric, dielectric, and elastic properties are developed to provide some insight into the nonlinear behavior of ferroelectric ceramics.

II THEORETICAL

Based on the model by Arlt et al.[5], we choose primed and unprimed coordinate systems on a basic piezoelectric element, which contains a single 90° domain wall, as shown in Fig.1. Δl is the displacement of domain wall and **A** is the area of the vibrating domain wall per unit volume. A displacement Δl of the domain wall gives rise to a change in the electric dipole moment, δP_i, in volume $\Delta l \mathbf{A}$ [5,6],

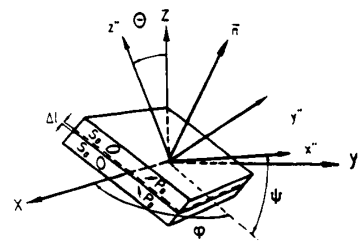

Fig.1. In 90⁰- domain in tetragonl ceramics, the relative orientation of unprimed and primed coordinate systems is characterized by angles (φ, ψ, θ) which are called the Eulerian angles.

$$\delta P_i = \Delta l\, A P_o \begin{bmatrix} f_1(\Theta,\Phi,\Psi) \\ f_2(\Theta,\Phi,\Psi) \\ f_3(\Theta,\Phi,\Psi) \end{bmatrix} \qquad (1)$$

where $f_1 = [\cos\Theta\sin\Phi\sin\Psi - \cos\Psi\cos\Phi]$,

$f_2 = -[\cos\Psi\sin\Phi + \cos\Theta\cos\Phi\sin\Psi]$

$f_3 = -\sin\Psi\sin\Theta$; and also induces a change in the elastic dipole moment,

$$\delta U_{ij} = \Delta l\, A S_o \begin{bmatrix} F_{11} & F_{12} & F_{13} \\ F_{12} & F_{22} & F_{23} \\ F_{13} & F_{23} & F_{33} \end{bmatrix} \qquad (2)$$

in which

$F_{11} = 2[\sin\Theta\cos\Psi\cos\Phi\sin\Phi - \cos\Theta\sin\Theta\sin^2\Phi\sin\Psi]$

$F_{22} = -2[\sin\Theta\cos\Psi\cos\Phi\sin\Phi + \cos\Theta\sin\Theta\cos^2\Phi\sin\Psi]$

$F_{33} = \cos\Theta\sin\Theta\sin\Psi$

$F_{23} = [\cos\Theta\cos\Psi\sin\Phi + \cos^2\Theta\sin\Psi\cos\Phi - \sin^2\Theta\sin\Psi\cos\Phi]$

$F_{13} = [\cos\Theta\cos\Psi\cos\Phi - \cos^2\Theta\sin\Psi\sin\Phi]$

$F_{12} = [\sin\Theta\cos\Psi\sin^2F + 2\sin\Theta\cos\Theta\sin\Psi\cos\Phi\sin\Phi - \sin\Theta\cos\Psi\cos^2\Phi]$

P_o and S_o are the spontaneous polarization and strain, respectively, in the tetragonal phase of the ferroelectric ceramics.

It has been shown [7] experimentally that the domain wall motion in poled ceramics is highly nonlinear. Therefore, the potential energy of the domain wall may be expanded as a polynomial function of the domain wall displacement:

$$U = U_o + A_1 \Delta l^2 + A_2 \Delta l^3 + A_3 \Delta l^4 + \text{higher- order terms} \quad (3)$$

where U_o is the thermodynamic energy of a domain boundary, which is assumed to be independent of domain wall motion. The presence of the third power term describes the asymmetric feature of the domain wall motion in a poled ceramic.

Therefore, the differential equation of the forced

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vibration a 90^0 domain wall in a poled ceramic may be expressed as follows:

$$Am\ddot{\Delta l} + Ab\dot{\Delta l} + \frac{\partial U}{\partial \Delta l} = -\left(\frac{\partial W_E}{\partial \Delta l} + \frac{\partial W_M}{\partial \Delta l}\right) \quad (4)$$

where m represents the effective mass of the domain wall, b is the damping constant, and W_E and W_M are the energies of the dipoles δP_i and δU_{ij}, respectively. The physical origin of the restoring force and damping are not discussed here.

According to Fousek[7], the resonance frequency of domain walls would be much higher than our measuring frequencies which are lower than 100 MHz. Thus, the term of inertia can be neglected. This means such a strong damping is assumed that an equation of relaxation with samll second- and third-order terms remains:

$$Ab\dot{\Delta l} + 2A\,C_1\,\Delta l + AC_2\,\Delta l^2 + AC_3\,\Delta l^3 = -\left(\frac{\partial W_E}{\partial \Delta l} + \frac{\partial W_M}{\partial \Delta l}\right) \quad (4')$$

This equation implies that the nature of the extrinsic nonlinear properties in ferroelectric ceramics is supposed to lie in the dependence between the amplitude of displacement and the stiffness of the material with respect to the damped motion of 90^0 domain walls.

The condition in Eq.(4')

$$C_1\Delta l >> C_2\Delta l^2,\ C_3\Delta l^3$$

allows one to employ a perturbation method of solution.

In order to obtain the induced polarization δP_i and strain δU_{ij} in domain wall motions, one may approximately solve Eq.(4') by using perturbation method[8].

III. LINEAR AND NONLINEAR COEFFICIENTS

We now only consider the linear and nonlinear dielectric, piezoelectric, and elastic properties, which are caused by 90^0 domain wall motions in ferroelectric ceramics. The average induced polarization and strain can be calculated by integrating δU_{ij} and δP_i over all domain wall orientations in the sample, as shown in Fig.2.

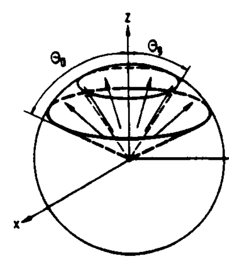

Fig.2. Simplest distribution of the orientation of the basic cells with respect to the poling axis. After Arlt et al.[5](1987).

For the average induced polarization ΔP_i, and the induced strain ΔU_{ij} in ferroelectric ceramics, the approximate solutions up to the second order may be expressed as:

$$\Delta U_j = [\Delta d_{jk} + \Delta r_{jki} E_i]E_k + [\Delta S_{ij}\,\Delta C_{ijm} T_m]T_j + \Delta e_{jkm} E_k T_m$$

$$i, j, m = 1, 2, 3, 4, 5, 6; \quad k, n = 1, 2, 3$$

$$\Delta P_n = [\Delta\varepsilon_{nk} + \Delta Q_{npk} E_p]E_k + [\Delta d_{nj} + \Delta e_{njm} T_m]T_j + \Delta r_{npj} E_p T_j$$

$$p, n, k = 1, 2, 3, 4, 5, 6; \quad j, m = 1, 2, 3 \quad (5)$$

Where ΔP_n is the induced polarization; ΔU_j is the strain component in Vogt notation; T_j is the stress, and E_k is the electric field.

The values Δd_{ik}, Δs_{ij}, and Δe_{nk} are linear piezoelectric, elastic, and dielectric parameters:

$$\Delta d_{kj} = \int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} S_o P_o\, K(\omega) f_k F_j\, Z(\Theta) d\Theta$$

$$\Delta s_{ij} = \int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} S_o^2\, K(\omega) F_i F_j\, Z(\Theta) d\Theta \quad (6)$$

$$\Delta \varepsilon_{kl} = \int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} P_o^2\, K(\omega) f_k f_l\, Z(\Theta) d\Theta$$

where $F_1=F_{11}$; $F_2=F_{22}$; $F_3=F_{33}$; $F_4=F_{23}$; $F_5=F_{13}$; $F_6=F_{12}$

$$K(\omega, C, A) = \int_0^{\infty} \frac{Ag(\tau)d\tau}{4C_1(1+j\omega\tau)}$$

$g(\tau)$ is the relaxation time distribution[5] (if there exist more than one relaxation time); $Z(\Theta)$ is the certain distribution function. All quantities having the Δ symbol are caused by the 90^0-domain wall vibration only.

Finally, we may get the following matrixes:

$$\Delta d_{ij} = \begin{bmatrix} 0 & 0 & \Delta d_{31} \\ 0 & 0 & \Delta d_{31} \\ 0 & 0 & \Delta d_{33} \\ 0 & \Delta d_{15} & 0 \\ \Delta d_{15} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}; \quad \Delta \varepsilon_{ij} = \begin{bmatrix} \Delta\varepsilon_{11} & 0 & 0 \\ 0 & \Delta\varepsilon_{11} & 0 \\ 0 & 0 & \Delta\varepsilon_{33} \end{bmatrix}$$

$$\Delta s_{ij} = \begin{bmatrix} \Delta s_{11} & \Delta s_{12} & \Delta s_{13} & & & \\ \Delta s_{12} & \Delta s_{11} & \Delta s_{13} & & \mathrm{O} & \\ \Delta s_{31} & \Delta s_{31} & \Delta s_{33} & & & \\ & & & \Delta s_{44} & 0 & 0 \\ & \mathrm{O} & & 0 & \Delta s_{44} & 0 \\ & & & 0 & 0 & \Delta s_{66} \end{bmatrix} \quad (7)$$

Δr_{jki} (electrostrictive coefficients) and ΔQ_{lki} (nonlinear dielectric coefficients) can be expressed as follows:

$$\Delta r_{j\,ki} = \int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} 2S_o\, P_o\, K'(\omega)\, F_j\, f_k\, f_i\, Z(\Theta)\, d\Theta$$

$$\Delta Q_{lki} = \int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} P_o^3\, K'(\omega) f_l\, f_i\, f_k\, Z(\Theta)\, d\Theta \quad (8)$$

$$K'(\omega, C, A) = \int_0^{\infty} \frac{C_2\, Ag(\tau)d\tau}{[4C_1(1+j\omega\tau)]^3}$$

The electroacoustic coefficients Δe_{ijm} (the coefficients of the nonlinear piezoelectric effect) describe the change in the velocity of elastic waves, and they can be given by:

$$\Delta e_{ijm}=\int_0^{2\pi} d\varphi \int_0^{2\pi} d\phi \int_0^{\pi} 2P_0 S_0^2\ Z(\theta)\ K'f_i F_j F_m\, d\theta \qquad (9)$$

$$i = 1, 2, 3.\ ; \qquad j;\ m = 1, 2, 3, 4, 5, 6.$$

It is believed extrinsic contribution to the electroacoustic effect may be two orders of magnitude larger than the intrinsic contribution in some single crystals[4].

The third order elastic coefficients are:

$$\Delta C_{ijm}=\int_0^{2\pi} d\varphi \int_0^{2\pi} d\phi \int_0^{\pi} 2S_0^3\ Z(\theta)\ K'F_i F_j F_m\, d\theta \qquad (10)$$

where: i, j, m = 1, 2, 3, 4, 5, 6. There are twenty one non-vanishing coefficients.

In the third approximation, the expressions for the higher order nonlinear piezoelectric coefficients ΔG_{ilki} and dielectric coefficients ΔH_{lkim} can be written by the following integrals:

$$[\Delta G_{iklj}]=\int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} S_0 P_0^4\ K''(\omega)\ F_j\, f_i\ f_k\ f_l\ Z(\Theta) d\Theta$$

$$[\Delta H_{lkim}]=\int_0^{2\pi} d\phi \int_0^{2\pi} d\psi \int_0^{\pi} P_0^5\ K''(\omega) f_l f_k\ f_i f_m\ Z(\Theta) d\Theta$$

$$K''(\omega,C,A) = \int_0^{\infty} \left\{\frac{4C_2^2 A^3 g(\tau)}{[4C_1(1+j\omega\tau)]^5} + \frac{2C_3 A^3 g(\tau)}{[4C_1(1+j\omega\tau)]^4}\right\} d\tau$$

If Ti= 0; the induced longitudinal strain due to domain wall motion is:

$$\Delta U_3 = \Delta d_{33} E_0 + \Delta r_{333} E_0^2 + \Delta G_{3333} E_0^3 \qquad (12)$$

From Eqs.(11) and (12), it can be found there are two factors that affect the magnitudes of the dielectric and piezoelectric coefficents: one is the area of the vibrating domain wall per unit volume; and the other is the presence of higher order harmornics.

Similarly, the shear strain under applied a.c. field can be expressed as:

$$\Delta U_5 = \Delta d_{15} E_0 + \Delta G_{1115} E_0^3 \qquad (13)$$

which indicates that there is no second harmonic in shear vibrations. Therefore, the dependence of Δd_{15} and $\Delta\varepsilon_{11}$ on alternating electric field should have different characteristics from those of Δd_{33}, Δd_{13}, and $\Delta\varepsilon_{33}$.

IV. EXPERIMENTAL RESULTS

Fig.3. shows the piezoelectric constants d_{ij} and the dielectric constants ε_{ii} as a function of applied a.c. field strength. When the magnitude of applied electric field is below a certain limiting value E_t (called the threshold field $E<E_t$), d_{ij} and ε_{ii} remain constant.. But beyond threshold field ($E > E_t$) dij and eij increase with the amplitude of a.c. electric field. Since the threshold point is not always clearly defined, we must use $\Delta d/d$ ($\Delta\varepsilon/\varepsilon$)> 5% as the criteria. For different materials, frequencies and temperatures, there are different threshold fields.

For instance, for PZT-5 with F= 200 Hz and T=25°C, the threshold field is about 300 V/cm.

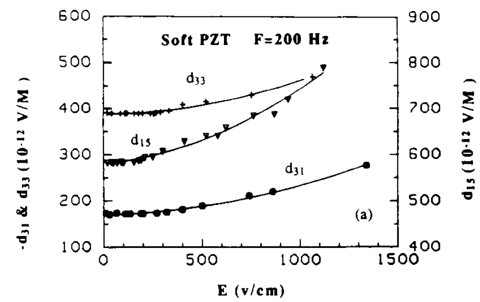

Fig.3. (a). A.C electric field dependence of piezoelectric coefficients for d_{31}, d_{33}, and d_{15}.

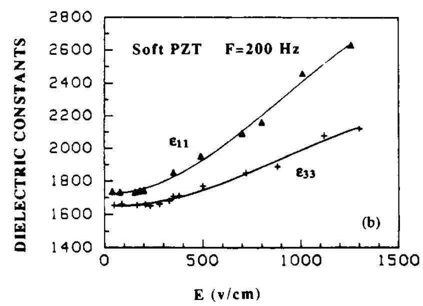

Fig.3.(b).A.C. electric field dependence of dielectric coefficients ε_{11}, and ε_{33}

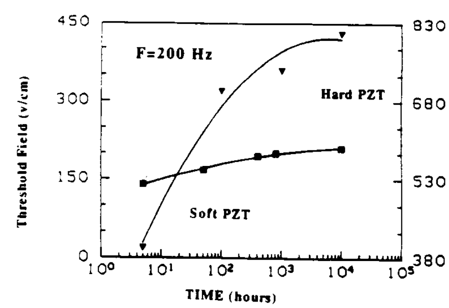

Fig.4. The threshold field of piezoelectric coeffcients d_{33} for both poled soft and hard PZT ceramics as a function of aging time.

Fig.4. shows the threshold fields of piezoelectric coefficients increase with increasing aging time. It is widely accepted that reductions of the non180°-domain wall mobility and area are responsible for the aging phenomena in ferroelectric ceramics. This indicates that the threshold field is closely related to non180°-domain wall motion. The bias field and temperature dependence of the threshold field are shown in Fig.5. Similarly, as a d.c. bias favours coalescence, lowering the temperature has the same effect as an applied positive d.c. bias. By using an oscilloscope to follow the polarization (P) and strain (S) versus electric field (E) at a fixed frequency, it was found that the P and S vs E curves are straight lines at small oscillation amplitudes. When the amplitude of the a.c. field increases, consequently, the average slope of the hysteresis increases. This indicates that nonlinear vibration of domain wall seems to appear because macrohysteresis effects originate from the lossy reversal of the spontaneous polarization of ferroelectric domains.

It is obvious that the shape of hysteresis loop varies with different directions of the a.c. field with respect to

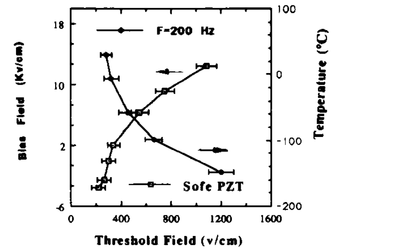

Fig.5. The threshold field of dielectric constant ε_{33} for poled PZT ceramics as a function of positive d.c. electric bias field, and the threshold field of dielectric coefficient ε_{33} versus temperature.

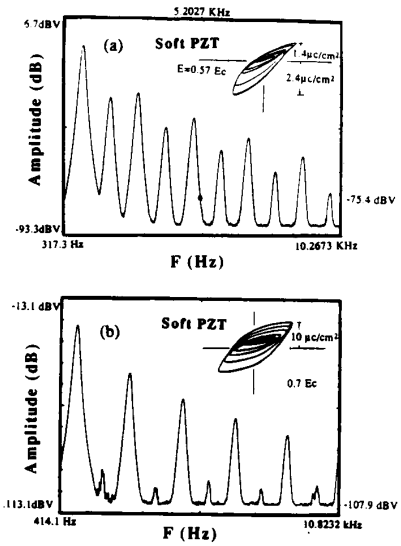

Fig.6. Spectral analyses of the output voltage of dielectric respones demonstrating the presence of higher harmonic components in the signal
(a) Electric field parallel to the poling direction; the amplitude of a.c. field 0.6 Ec
(b) Electric field perpendicular to the poling direction

poling axis of the sample. When the direction of a.c. field is perpendicular to the polarization direction of the sample, the shape of the hysterisis loop is always symmetric. Conversely, if the direction of a.c. field is parallel to the polarization direction of the poled sample, the shape of hysterisis loop is no longer symmetric at certain ranges of external field strength. Examining the spectrum of the dielectric and piezoelectric responses in PZT ceramics, we found that when an a. c. electric field E is parallel to the poling direction, there exist both odd and even harmornics at certain amplitudes of external field. By constrast, when $E \perp P$, there exist only odd harmonics, as shown in Figs.6. These results are consistent with the theoretical analysis presented earlier.

Fig.7. shows the dependence of relative dielectric permittivities ε_{33} & ε_{11} versus temperature at different frequencies measured using a HP4270A automatic capacitance bridge. eii increased gradually over the temperature span -160 °C to 50 °C.

It was found that difference values between ε_{11} and ε_{33} gradually decrease with increasing frequency. This may be attributed to the fact that there exists different types of excitation of non-180° domain walls . In other words, these indicate that parts of the difference between the values of ε_{11} and ε_{33} originate from extrinsic

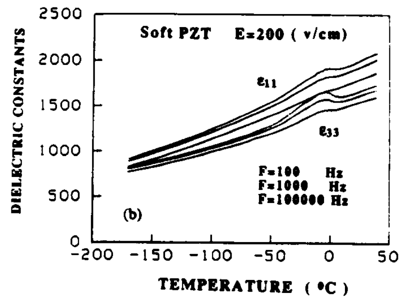

Fig.7. The ε_{11} and ε_{33} as a function of temperature for soft PZT at different frequencies

properties, since 90°-domain wall motion is inherently a lossy process[9].

In short, all experimental results presented above shows that in ferroelectric ceramics, under certain circumstances, electromechanical nonlinearity originates from the nonlinear vibration of non-180° domain wall motions and movement of interphase boundaries in ferroelectric ceramic.

V. SUMMARY

A series of measurements is presented in an attempt to characterize the electromechanical dynamic response of PZT piezocéramics which shows that electromechanical nonlinearity in ferroelectric ceramics is closely associated with domain wall motions. A tentative phenomenological model has been proposed, which may be used to evaluate the macroscopic nonlinear parameters due to 90°-domain wall vibrations in ferroelectric ceramics. The theoretical description appears to give qualitative agreement with the experimental results.

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