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Frequency and Field-intensity Dependence of Dielectric and Piezoelectric Responses of Oxide Ferroelectrics
酸化物強誘電体の誘電・圧電応答の周波数および電界強度依存性
--- Description of Nonlinear Response and Dissipation Effect in Piezoelectric Fundamental Equations ----
圧電基本式における非線形応答および損失の記述

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1. Introduction

Promising applications of piezoelectric ceramics have been changing from devices operated under low electric fields in electronic circuits to those driven by high electric fields such as actuators for ink-jet heads, fuel injectors, ultrasonic transducers etc. Under such large-signal operations, the piezoelectric, dielectric, and elastic responses of piezoelectric ceramics deviate from the conventional linear piezoelectric theory, leading to the appearance of several nonlinear phenomena which causes the unstable operation and heat generation of the devices. Thus, the design of promising piezoelectric applications highly requires the piezoelectric theory including the nonlinear and dissipation effects. However, the theory has not been established yet and the nonlinear phenomena have not been fully understood. The authors have been studying piezoelectric response of various materials to improve this situation, and recently, they have reached the goal of their purpose. In this presentation, their achievements will be summarized.

2. Assumption for new equations

The piezoelectric fundamental equations of d-form is given by

$$D = \varepsilon^{T}E + dT,\ S = s^{E}T + dE \qquad (1)$$

where D is electric displacement, ε^{T} is dielectric permittivity under a fixed stress, E is electric field, d is piezoelectric constant, T is stress, S is strain, s^{E} is elastic compliance under a fixed E-field. Incorporation of nonlinear effect into Eq. (1) demands the series expansion in terms of E and T, and the incorporation of dissipation factor demands the conversion of real material constants to complex ones. These changes obviously results too complicated equations to solve. For handling nonlinear response and dissipation effect in piezoelectric fundamental equations, we must employ some assumptions to simplify the equations. After a long time study, we have found the assumption was that the polarization and the strain should be proportional without phase lag. Electric displacement and strain induced by piezoelectric effect can be written as

$$D = eS, \qquad S = gD \qquad (2)$$

where e and g are piezoelectric constants. Polarization P can be regarded as electric displacement D in piezoelectric ceramics with high permittivity. Thus, our assumption that the polarization and the strain should be proportional without phase lag can be described as piezoelectric e and g-constants are real numbers. After analysis and consideration, we finally concluded the real meaning of the assumption could be simplified as the electrostrictive Q constant was a real number.

There was a very big obstacle to accept the assumption in Eq. (2) in our study, which was the relationship between dielectric, elastic and piezoelectric losses. If the e and g-constants in Eq. (2) are real numbers, the dielectric loss and elastic loss must be identical, but we know that the both losses are independent from various experimental results. In order to avoid this discrepancy, we had to accept that the loss of d-constant was different between the direct and the converse effect. The fundamental equation of d-form (Eq. (1)) should be revised to incorporate dissipation effect as follows:

$$\begin{gathered} D = \varepsilon^{T^*}E + d^{[D]^*}T \\ S = s^{E^*}T + d^{[C]^*}E \\ \frac{d''^{[D]}}{d'^{[D]}} = \frac{s''^{E}}{s'^{E}} = \frac{1}{Q_m} \qquad \frac{d''^{[C]}}{d'^{[C]}} = \frac{\varepsilon''^{E}}{\varepsilon'^{E}} = \tan\delta \end{gathered} \qquad (3)$$

where $d^{[D]*}$ and $d^{[C]*}$ are complex d-constants in direct and converse effect, respectively, and Q_m is a mechanical quality factor. The above equations demand that d-constants in the direct effect and the converse effect are different. There is no intrinsic piezoelectric loss and the loss of d-constant in direct effect is equal to the loss of elastic constant, while that in converse effect is equal to the dielectric loss. This relationship has been experimentally confirmed.

3. Derivation of Fundamental Equations

Devonshire's theory was employed to derive the new piezoelectric fundamental equations including nonlinear response and dissipation effect. The Taylor series expansion of electric Gibbs energy $G_2(S, E)$ can be written as follows:

$$G_2(S,E)=\sum_n \frac{1}{n!}\left(S\frac{\partial}{\partial S}+T\frac{\partial}{\partial E}\right)^n G_2{}^n \qquad (4)$$

Taking up to the fourth-order terms, we obtain

$$\begin{aligned}G_2(S,E)=&\frac{1}{2}\left(\frac{\partial^2 G_2}{\partial S^2}\right)_E S^2+\left(\frac{\partial^2 G_2}{\partial S\partial E}\right)SE+\frac{1}{2}\left(\frac{\partial^2 G_2}{\partial E^2}\right)_S E+\frac{1}{2}\left(\frac{\partial^3 G_2}{\partial S\partial E^2}\right)SE^2\\&+\frac{1}{6}\left(\frac{\partial^3 G_2}{\partial S^3}\right)_E S^3+\frac{1}{24}\left(\frac{\partial^4 G_2}{\partial S^4}\right)_E S^4+\frac{1}{6}\left(\frac{\partial^3 G_2}{\partial E^3}\right)_S E^3+\frac{1}{24}\left(\frac{\partial^4 G_2}{\partial E^4}\right)_S E^4 \qquad (5)\\&+\frac{1}{2}\left(\frac{\partial^3 G_2}{\partial S^2\partial E}\right)S^2E+\frac{1}{6}\left(\frac{\partial^4 G_2}{\partial S^3\partial E}\right)S^3E+\frac{1}{4}\left(\frac{\partial^4 G_2}{\partial S^2\partial E^2}\right)S^2E^2+\frac{1}{6}\left(\frac{\partial^4 G_2}{\partial S\partial E^3}\right)SE^3.\end{aligned}$$

The terms in the third line in the above equation are all zero because piezoelectric e-constant given by differentiating G_2 in terms of S followed by E is constant and independent of S or E from the assumption. Then, the free energy can be simplified as:

$$\begin{aligned}G_2(S,E)=&\frac{1}{2}c^{E(1)}S^2+\frac{1}{3}c^{E(2)}S^3+\frac{1}{4}c^{E(3)}S^4\\&-eSE-\gamma SE^2-\frac{1}{2}\varepsilon^{S(1)}E-\frac{1}{3}\varepsilon^{S(2)}E^3-\frac{1}{4}\varepsilon^{S(3)}E^4. \qquad (6)\end{aligned}$$

where $c^{E(1)}$, $c^{E(2)}$ and $c^{E(3)}$ are first-, second- and third-order elastic stiffness under fixed E-field, γ is electrostrictive coefficient, $e^{S(1)}$, $e^{S(2)}$, $e^{S(3)}$ are first-, second-and third-order dielectric permittivity under fixed strain. By differentiating above free energy G_2 in terms of S or E, we obtain a set of equation for e-form as follows:

$$\begin{cases}T=c^{E(1)^*}S+c^{E(2)^*}S^2+c^{E(3)^*}S^3-eE-\gamma E^2\\ D=eS+2\gamma ES+\varepsilon^{S(1)^*}E+\varepsilon^{S(2)^*}E^2+\varepsilon^{S(3)^*}E^3\end{cases} \qquad (7)$$

This is the piezoelectric fundamental equation of e-form and other forms can be derived in the same way. It should be noted that the above equations include all terms that we need in any analyses.

4. Verification of new equations

New fundamental eqations were experimentally verified. An example is a wave form of vibration velocity of PZT, PZT+PMN and PMN driven under ac-field. The wave forms are shown in **Fig. 1**. The vibration velocity was calculated from Eq. (7) combined with the eqauation of motion by igonoring higher orfer terms in Eq. (7). Very good fitting was obtained even for PMN-PT showing mixed response of piezoelectric and electrostrictive effects. Another example is the jumping phenomenon observed for Mn-doped PZT (**Fig. 2**). Asymmetrical peaks of vibration velocity due to the nonlinear response were completely reproduced by the calculation with ignoring electrostrictive coefficient in Eq. (7). We found that the jumping phenomenon was due to nonlinear elastic constants.

5. Summary

We have derived a new piezoelectric fundamental equations including nonlinear response and dissipation effect. The assumtion we used was that the electrostrictive Q constant was a real numbers. The equations sucessfully explain the nonlinear piezoelectric response as well as electrostrictive effect.

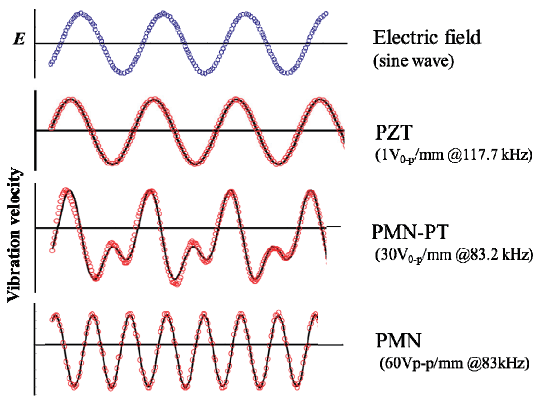

Fig. 1 Wave forms of vibration velocity of PZT, PZT+PMN and PMN. Solid lines are calculated using new equations.

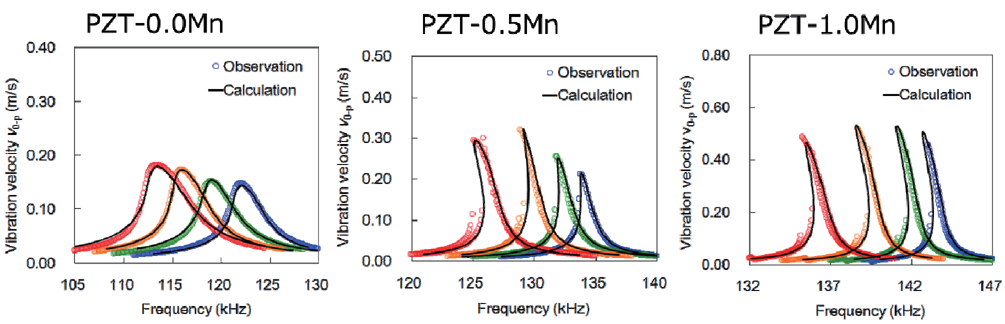

Fig. 2 Vibration velocity around resonance frequency measured for PZT added with MnO.