Measurement of the Nonlinear Coefficient of a Second-Higher Term in Lead Zirconate Titanate Piezoelectric Ceramics with Half-Surface Electrodes

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半面電極試料を用いた PZT 圧電セラミックスの 2 次項非線形定数測定

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Second-harmonic voltage between a pair of electrodes, appearing due to nonlinearity, was theoretically calculated in piezoelectric rectangular vibrators with a pair of half electrodes which were driven by a sinusoidal constant current having the resonance frequency of length-extensional $1/2\lambda$ -mode vibration. The theoretical calculation was performed using the electrical equivalent circuit of a piezoelectric rectangular vibrator. A material constant as a nonlinear coefficient of second-higher term can be derived by using an equation, as that obtained in the present paper, showing the relationship between the magnitude of second harmonic voltage and the nonlinear coefficient of second-higher term. [Received October 26, 2004; Accepted January 20, 2005]

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1. Introduction

Nonlinear phenomena have become serious problems that prevent operation stability and high efficiency in piezoelectric ceramics used in electronic devices for high-power applications, because of the downscaling of electronic devices and increase of their power density. The current jump phenomenon and harmonic voltage generations are typical nonlinear phenomena that are observed in piezoelectric vibrators driven with a high power at around the resonance frequency.^{1)–4)}

In order to quantitatively analyze the nonlinearity in piezoelectric ceramics, it is necessary to obtain nonlinear coefficients. Although several researchers have published papers on nonlinear coefficients of lead zirconate titanate (PZT), since the frequencies were less than 100 Hz in those studies,^{5)–8)} it is difficult to apply those results directly to piezoelectric ceramics driven at the resonance frequency.

On the other hand, several researchers have analyzed the nonlinear phenomena in high-power vibrations in terms of the change of linear constants, from the viewpoint of practical use.^{9)–12)} However, since these analyses focused on the change of linear constants, it is necessarily impossible to define the nonlinear material constants that quantitatively represent nonlinearity.

In contrast to the studies on nonlinearity mentioned above, the principal objectives of our study are to define nonlinear material constants that quantitatively represent nonlinearity and to introduce the equation of the relationship between voltage and current representing quantitatively nonlinear phenomena arising with high-power vibrations. We already reported the theoretical analysis about the relationship between the nonlinear coefficient of a third-higher term, ξ_{D31} , and the third-harmonic voltage generating between electrodes when a rectangular ceramic with whole electrodes on both upper and lower surfaces was driven by a sinusoidal constant current having the resonance frequency of length-extensional $1/2\lambda$ -mode vibration.¹³⁾

The ξ_{D31} value rules over the current jump phenomenon which is one of the typical nonlinear phenomena. It is there-

fore possible to theoretically calculate the driving conditions causing the current jump phenomenon, if a ξ_{D31} value is obtained.¹⁴⁾ Because of these circumstances, we have investigated about the nonlinear coefficient of a third-higher term. On the other hand, the nonlinear coefficient of a second-higher term rules over the generation of the second harmonic voltage which is one of the nonlinear voltages. Since the second harmonic voltage has twice frequency of driving frequency, when a rectangular bar vibrator with half electrodes was driven in high power levels at resonance frequency, the frequency of the second harmonic voltage generating in the sample belong consequently in the frequency range of the second harmonic resonance. It is therefore anticipated that the influence of the second harmonic voltage becomes remarkable in the sample with half electrodes. Because of these circumstances, it is necessary to establish the measurement method also about the nonlinear-coefficient of second higher term. In the present study, we analyze the relationship between the second harmonic voltage and the nonlinear coefficient of second-higher term, using the electrical equivalent circuit, in a piezoelectric rectangular bar with a pair of half electrodes on both upper and lower surfaces. The nonlinear coefficient of second-higher term can be successively derived as a material constant in this analysis.

2. Nonlinear piezoelectric equation

As the nonlinear piezoelectric equations of a rectangular vibrator with whole surface electrodes as shown in Fig. 1 (a), Eqs. (1) and (2) were proposed in the previous paper.¹³⁾

$$E_3 = -\frac{g_{31}}{s_{11}^D} S_1 + \beta_{33}^S D_3 + \gamma_{D31} D_3^2 + \xi_{D31} D_3^3 \quad (1)$$

$$T_1 = \frac{1}{s_{11}^D} S_1 - \frac{g_{31}}{s_{11}^D} D_3 + \chi_{D31} D_3^2 + \zeta_{D31} D_3^3 \quad (2)$$

Since Eqs. (1) and (2) are nonlinear piezoelectric equations, it is difficult to obtain the relationship between voltage and current by solving exactly them. Equations (1) and (2) were

Fig. 1. Configuration of the rectangular bar vibrators ($l=29.5$ mm, $w=2.62\text{--}5.93$ mm, $b=0.9$ mm) with whole surface electrodes (a) and half electrodes (b).

therefore solved using an approximation as follows. The relationship between linear terms is independent even when nonlinear phenomena occur. Nonlinear voltages occurring in the sample can simply be added to the voltage of linear terms. This is the superposition between linear and nonlinear terms, as well as the perturbation of nonlinear terms to linear ones. That is, this is an approximation by perturbation, which is applied when nonlinear terms are small compared with linear terms.

Because of driving at around the resonance frequency, since the current flowing through the damping capacitance, C_s , is very small compared with that flowing through the motional impedance, the current flowing through C_s can be neglected. By adding nonlinear terms to the solution of the linear terms according to the approximation by perturbation mentioned above, we obtain

$$v_3 = L_1 \frac{di_3}{dt} + R_1 i_3 + \frac{1}{C_1} \int i_3 dt + \gamma'_{D31} \left(\int i_3 dt \right)^2 + \xi'_{D31} \left(\int i_3 dt \right)^3, \quad (3)$$

where v_3 is the voltage between electrodes, i_3 is the current, L_1 , C_1 and R_1 are equivalent-circuit constants, and γ'_{D31} and ξ'_{D31} are effective nonlinear coefficients for the vibrator. The effective nonlinear coefficients are constants for each vibrator, and change with the size of the vibrator.

When the sample is driven with a constant current of $i_3 = I_3 \sin \omega t$ at the resonance frequency, the voltage between electrodes, v_3 , is obtained by substituting $i_3 = I_3 \sin \omega t$ into Eq. (3).

$$v_3 = \left(\omega L_1 - \frac{1}{\omega C_1} \right) I_3 \cos \omega t + R_1 I_3 \sin \omega t + \frac{\gamma'_{D31}}{\omega^2} I_3^2 \cos^2 \omega t - \frac{\xi'_{D31}}{\omega^3} I_3^3 \cos^3 \omega t \quad (4)$$

Expanding $\cos^2 \omega t$ and $\cos^3 \omega t$ using trigonometric formulae for double and triple angles, Eq. (4) is rewritten as

$$v_3 = \frac{\gamma'_{D31}}{2\omega^2} I_3^2$$

$$+ \left(\omega L_1 - \frac{1}{\omega C_1} \right) I_3 \cos \omega t + R_1 I_3 \sin \omega t - \frac{3\xi'_{D31}}{4\omega^3} I_3^3 \cos \omega t + \frac{\gamma'_{D31}}{2\omega^2} I_3^2 \cos 2\omega t - \frac{\xi'_{D31}}{4\omega^3} I_3^3 \cos 3\omega t. \quad (5)$$

A dc component and 2ω component due to γ'_{D31} , and a ω component and 3ω component due to ξ'_{D31} , are generated as nonlinear voltages, respectively. We have quantitatively expressed nonlinear phenomena using Eq. (5).^{1)–4)}

3. Experimental procedures

In the measurement of the nonlinear coefficient of third-higher term, ξ_{D31} , a sample with whole electrodes was used in Fig. 1(a). When the sample is driven with a constant current at the fundamental resonance angular frequency ($\omega_r = 2\pi f_r$), the angular frequency of the third-harmonic voltage appearing between electrodes due to ξ_{D31} is $3\omega_r$. Since the sample with whole electrodes has many resonance frequencies at odd number times of fundamental resonance frequency, the third-harmonic resonance occurs at around $3\omega_r$. The magnitude of third-harmonic voltage appearing between electrodes was significantly influenced by the position of $3\omega_r$ on the third-harmonic resonance curve. The magnitude of third-harmonic voltage appearing between electrodes showed the maximum when $3\omega_r$ coincided exactly with the anti-resonance angular frequency ($\omega_{3a} = 2\pi f_{3a}$) of the third-harmonic resonance. Thus, the motional impedance at $3\omega_r$ was essential to calculate ξ_{D31} value from the magnitude of third-harmonic voltage.¹³⁾

Analogizing from the theoretical analysis used in derivation of ξ_{D31} , it is necessary that a piezoelectric resonance exists at around $2\omega_r$ to derive the nonlinear coefficient of second-higher term, γ_{D31} , from the magnitude of second-harmonic voltage. Since a sample with half electrodes shown in Fig. 1(b) has a piezoelectric resonance at around $2\omega_r$, the sample with half electrodes was used in the estimation of γ_{D31} .

The composition of the prepared samples is $0.95\text{Pb}(\text{Zr}_{0.51}\text{Ti}_{0.49})\text{O}_3\text{--}0.05\text{Pb}(\text{Sb}_{0.5}\text{Nb}_{0.5})\text{O}_3\text{--}0.015\text{MnO}$. The samples were prepared using oxide powders as starting materials, by means of a conventional method, namely, mixing by wet ball-milling, calcining at 850°C for 2 h in air, grinding by wet ball-milling, pressing at 100 MPa, and firing at 1240°C for 2 h in air. After firing silver electrodes onto the upper and lower half surfaces of the rectangular bars as shown in Fig. 1(b), the samples were poled under an electric field of 3 kV/mm for 10 min in a silicon oil bath kept at 120°C . Mechanical quality factor, Q_m , electromechanical coupling factor, k_{31} , piezoelectric d_{31} and h_{31} constants, elastic compliance, s_{11}^E , and relative permittivity, $\epsilon_{33}^T/\epsilon_0$, are listed in **Table 1**. These material constants were derived from the resonance-antiresonance characteristic of a whole electrodes sample in Fig. 1(a) measured at a low current level of less than 1 mA using an LF impedance analyzer (Hewlett-Packard, 4192A).

The second-harmonic voltage was measured using the constant-current circuit shown in **Fig. 2**. In constant-current driving, the sample should be driven by a sinusoidal current without any distortion. A power amplifier (NF Electronics Instruments, 4020) generates a second-harmonic voltage with a magnitude of 0.1 to 0.4% of the fundamental voltage; driving current consequently imposes a distortion via the second-harmonic voltage. In order to prevent the application of the second-harmonic voltage to the sample, a parallel resonance

Table 1. Piezoelectric Constants of the Sample

Q_m	1550
k_{31}	0.366
d_{31} ($\times 10^{-12}$ m/V)	121
h_{31} ($\times 10^9$ N/C)	1.27
s_{11}^E ($\times 10^{-12}$ Pa $^{-1}$)	11.4
$\varepsilon_{33}^T / \varepsilon_0$	1270

Fig. 2. Measuring circuit used for the constant-current driving.

circuit was inserted in series with the sample, as shown in Fig. 2. The resonance frequency of the parallel resonance circuit was adjusted to a frequency equal to two times the driving frequency. Since the parallel resonance circuit has an impedance above 100 kΩ at the frequency of the second-harmonic voltage of the power amplifier (equal to the frequency of the second-harmonic voltage generated in the sample), the parallel resonance circuit prevents the application of the second-harmonic voltage generated in the power amplifier to the sample, and simultaneously enables us to take the second-harmonic voltage, generating in the sample, out of the sample.

The series resistors, R_a and R_b , were used for the realization of constant-current driving and for monitoring the current. The current was kept constant by the series resistor R_a of 3 kΩ during the measurement, because the resonance resistance of the samples was less than 20 Ω at the resonance frequency. A resistor of 50 Ω was used as R_b . The frequency of a function generator (Hewlett-Packard, 3323B) was fixed at a fundamental resonance frequency of the $1/2\lambda$ -mode of the sample in Fig. 1(b), and the signal of the function generator was amplified by a power amplifier (NF Electronics Instruments, 4020). A spectrum analyzer (Hewlett-Packard, ESA-L1500A) was used for the measurement of the magnitudes of fundamental and second-harmonic voltages between electrodes of the sample.

4. Results and discussion

The f_{2a} is an anti-resonance frequency in the second-harmonic resonance. The second-harmonic resonance curve in Fig. 3(a) is representative of the curves of six samples differing slightly in the relation between f_r and f_{2a} , namely, showing the curve on the basis of f_{2a} , it is showed as Fig. 3(a). Since six resonance curves almost override one another, it is showed for convenience with a curve being representative. The slight differences in the relation between f_r and f_{2a} were caused by the slight change of the size and configuration of samples. When the six samples with the configuration shown in Fig. 1(b) are driven at the fundamental resonance frequency by the constant-current method, the impedance at two times the driving frequency shows various values on the second-harmonic

Fig. 3. Impedance values of six samples at $2 \times f_r$ on the second-harmonic resonance curve which is representative of the curves of six samples for convenience (a), and the magnitudes of second-harmonic voltage between electrodes of the six samples (b). The second-harmonic resonance and second-harmonic antiresonance frequencies, f_r and f_{2a} , are about 117 kHz and 120 kHz, respectively.

Fig. 4. Equivalent circuit of a piezoelectric vibrator with half electrodes. The second-harmonic nonlinear voltage source is included.

resonance curve, as shown in Fig. 3(a). The plotted circles in Fig. 3(a) show where $2 \times f_r$ occupies on the second-harmonic resonance curve.

Figure 3(b) shows the magnitude of second-harmonic voltages, V_{h2ap} , between a pair of electrodes when these six samples were driven at the fundamental resonance frequency by the constant current of 10 mA_{0-P}. When $2 \times f_r$ agreed with f_{2a} , the maximum V_{h2ap} was measured. This fact is the same circumstances as the third-harmonic voltage generating in the whole electrodes sample in Fig. 1(a).¹³⁾ The electrical equivalent circuit including nonlinear term of a piezoelectric ceramic can be therefore composed of motional impedance parameters originating in fundamental and second-harmonic resonances (L_1 , C_1 , R_1 , L_2 , C_2 , R_2), damping capacitance, C_s , and the nonlinear voltage source v_{731} . Figure 4 is the equivalent circuit including v_{731} .

The magnitude of second-harmonic voltage appearing between electrodes, V_{h2ap} , can be expressed as Eq. (6) by con-

sidering the equivalent circuit and the magnitude of the 2ω component in Eq. (5).

$$V_{h2ap} = \frac{\gamma'_{D31} I_3^2}{2\omega_r^2} \cdot \frac{Z_{CS2}}{Z_{CS2} + Z_{m2}} \quad (6)$$

Here Z_{CS2} and Z_{m2} are the impedance of damping capacitance, C_s , and the second motional impedance at $2\omega_r$. Since Z_{CS2} and Z_{m2} can be expressed as Eqs. (7) and (8), Eq. (6) is rewritten as Eq. (9).

$$Z_{CS2} = \frac{1}{j2\omega_r C_s} \quad (7)$$

$$Z_{m2} = R_2 + j \left(2\omega_r L_2 - \frac{1}{2\omega_r C_2} \right) \quad (8)$$

$$V_{h2ap} = \frac{\gamma'_{D31} I_3^2}{2\omega_r^2} \cdot \frac{1}{1 - 2\omega_r C_s \left(2\omega_r L_2 - \frac{1}{2\omega_r C_2} \right) + j2\omega_r C_s R_2} \quad (9)$$

Equation (9) shows that the V_{h2ap} depends greatly on where the frequency of $2\omega_r$ is on the second harmonic resonance curve. When $2\omega_r$ equals $\omega_{2a} = 2\pi f_{2a}$, it is expressed as

$$2\omega_r C_s \left(2\omega_r L_2 - \frac{1}{2\omega_r C_2} \right) = 1 \quad (10)$$

V_{h2ap} shows therefore the maximum value. The curve fitted to the results of Fig. 3(b), obtained by substituting the equivalent circuit constants of the second harmonic resonance of a sample used in the measurement of V_{h2ap} in Fig. 3(b) ($C_s = 759$ pF, $R_2 = 33.1$ Ω , $L_2 = 39.6$ mH, $C_2 = 46.4$ pF) and an adequate value of γ'_{D31} (6.5×10^{14} V/C²) into Eq. (9), is shown as a solid line in Fig. 3(b). The fitted curve agrees well with the measured values. The results are the same as the case of ξ_{D31} . Thus, by inserting the nonlinear voltage source, the change of V_{h2ap} can be quantitatively explained.

The samples different in width, w , and equal in length and thickness ($l = 29.5$ mm, $b = 0.9$ mm) of Fig. 1(b) were driven at each resonance angular frequency, ω_r , by the constant current method. The magnitudes of second-harmonic voltage appearing between electrodes, V_{h2ap} , were measured at various driving currents, I_3 . **Figure 5** shows γ'_{D31} values obtained by substituting V_{h2ap} and I_3 values into Eq. (9). The horizontal axis is shown by average current densities ($2I_3/wl$). Since γ'_{D31} represents the quantitative relation between V_{h2ap} and I_3 as shown in Eq. (6), γ'_{D31} shows different values if electrode surface differs. Increasing electrode surface, decreasing γ'_{D31} value. The purpose of the paper is to theoretically analyze the quantitative relationship between the effective nonlinear coefficient, γ'_{D31} , and the nonlinear coefficient as a material constant, γ_{D31} .

5. Theoretical analysis

The current density and electric flux density in a rectangular bar are distributed with the cosine distribution in the length direction of the sample driven at the fundamental resonance angular frequency, ω_r , by the constant-current method. Expressing x -distance, where the origin is at the center of the sample, D_3 is expressed

$$D_3 = -D_0 \cos \frac{\pi}{l} x \cdot \cos \omega_r t, \quad (11)$$

where, x is restricted in the region of electrode ($0 \leq x \leq l/2$). The nonlinear voltage generated in the sample due to the

Fig. 5. Effective nonlinear coefficient of second-higher term γ'_{D31} as a function of average current density in the samples with different sample width.

second-higher term is expressed as

$$v_{\gamma31} = b\gamma_{D31}D_3^2 = b\gamma_{D31}D_0^2 \cos^2 \frac{\pi}{l} x \cdot \cos^2 \omega_r t, \quad (12)$$

where b is the thickness of the sample. Expressing current density as $j_3 = \partial D_3 / \partial t$, the sample current, i_3 , is expressed as

$$i_3 = w \int_0^{l/2} j_3 dx = \frac{lw\omega_r D_0}{\pi} \sin \omega_r t = I_3 \sin \omega_r t. \quad (13)$$

From Eq. (13), we obtain

$$D_0 = \frac{I_3 \pi}{lw\omega_r}. \quad (14)$$

Substituting Eq. (14) into Eq. (12), we get

$$\begin{aligned} v_{\gamma31} &= b\gamma_{D31} \frac{I_3^2 \pi^2}{l^2 w^2 \omega_r^2} \cos^2 \frac{\pi}{l} x \cdot \cos^2 \omega_r t \\ &= b\gamma_{D31} \frac{I_3^2 \pi^2}{4l^2 w^2 \omega_r^2} \left\{ 1 + \cos \frac{2\pi}{l} x + \left(1 + \cos \frac{2\pi}{l} x \right) \right. \\ &\quad \left. \times \cos 2\omega_r t \right\}. \end{aligned} \quad (15)$$

From Eq. (15), the magnitude of the second-harmonic voltage generating in the sample is expressed as

$$V_{h2}(x) = b\gamma_{D31} \frac{I_3^2 \pi^2}{4l^2 w^2 \omega_r^2} \left(1 + \cos \frac{2\pi}{l} x \right). \quad (16)$$

It is necessary to investigate how second-harmonic voltage is measured between electrodes when the second-harmonic voltage with cosine distribution generates in the sample having electrodes on the upper and lower surfaces. Since $2\omega_r$ is under the influence of second-harmonic piezoelectric resonance, the distribution of motional impedance consequently exists in the sample. The impedance against the second-harmonic voltage has reactance components since the second-harmonic resonance angular frequency, ω_{2r} , is different from $2\omega_r$. The impedance per meter in the length direction, $Z_{m2}(x)$, is expressed as

$$Z_{m2}(x) = Z_{m2}(l/4) \frac{1}{\sin \frac{2\pi}{l} x} \quad (17)$$

where $Z_{m2}(l/4)$ is the impedance per meter at $x = l/4$.

In the sample having distributions of both second-harmonic electromotive force and impedance, the voltage measured between electrodes can be calculated using Millman's theorem.¹⁵⁾

Fig. 6. Nonlinear coefficient of second-higher term γ_{D31} as a function of average current density in the samples with different sample width.

On the other hand, in the case of $2\omega_r \neq \omega_{2r}$, the influence of Z_{CS2} can be considered. Considering the influence of Z_{CS2} and using Millman's theorem, the magnitude of the second-harmonic voltage appearing between electrodes is expressed as

$$V_{h2ap} = \frac{\int_0^{l/2} \frac{V_{h2}(x)}{Z_{m2}(x)} dx}{\int_0^{l/2} \frac{1}{Z_{m2}(x)} dx} \cdot \frac{Z_{CS2}}{Z_{CS2} + Z_{m2}}$$

$$= b\gamma_{D31} \frac{I_3^2 \pi^2}{4l^2 w^2 \omega_r^2} \cdot \frac{Z_{CS2}}{Z_{CS2} + Z_{m2}} \quad (18)$$

Comparing Eq. (6) with Eq. (18), as the relation of γ_{D31} and γ'_{D31} , we get

$$\gamma_{D31} = \frac{2l^2 w^2}{b\pi^2} \gamma'_{D31} \quad (19)$$

By converting γ'_{D31} values in Fig. 5 into γ_{D31} values using Eq. (19), Fig. 6 is obtained. The γ_{D31} values converge with around a constant value ($\gamma_{D31} = 2.3 \times 10^9 \text{ m}^3 \text{ V/C}^2$). This fact demonstrates that the analysis in the present paper is valid.

6. Conclusions

When a piezoelectric rectangular bar with half electrodes was driven at the fundamental resonance frequency by the constant-current method, nonlinear second-harmonic voltage appearing between electrodes due to γ_{D31} are expressed as Eq. (18). The nonlinear coefficient of the second-higher term, γ_{D31} , can be therefore derived as a material constant by measuring the magnitude of the nonlinear second-harmonic voltage appearing between electrodes and using the value into Eq. (18).

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