Measurement of the Nonlinear Coefficient of a Third-Higher Term in Lead Zirconate Titanate Piezoelectric Ceramics

Shinjiro TASHIRO, Keisuke ISHII and Kunihiro NAGATA
Department of Materials Science and Engineering, The National Defense Academy, 1–10–20, Hashirimizu, Yokosuka-shi 239–8686

Abstract

Fundamental and 3rd-harmonic voltages between a pair of electrodes, appearing due to nonlinearity, were theoretically calculated in a piezoelectric rectangular vibrator which was driven by a sinusoidal constant current having the resonance frequency of length-extensional 1/22-mode vibration. The theoretical calculation was performed using the electrical equivalent circuit of a piezoelectric rectangular vibrator. Since the fundamental and 3rd-harmonic voltages were generated with cosine distributions in the sample and the motional impedance to each generated voltage also has a cosine distribution, Millman’s theorem was used in the analysis of the fundamental and 3rd-harmonic voltages between a pair of electrodes. This analysis clarifies quantitatively the relationship between the nonlinear voltages between electrodes and a nonlinear coefficient used as a material constant.

Key-words: Nonlinear piezoelectric coefficient, Lead zirconate titanate, High-power vibration, Piezoelectric vibrator

1. Introduction

Because of the downscaling of electronic devices and increase of their power density, nonlinear phenomena have become serious problems that prevent operation stability and high efficiency in piezoelectric ceramics used in electronic devices for high-power applications. 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.1–3

In order to quantitatively analyze the nonlinearity in piezoelectric ceramics, it is necessary to obtain nonlinear coefficients. In particular, the nonlinear coefficient of the 3rd-higher term (4th-order nonlinear coefficient) is essential in estimating the vibration condition at which the current jump phenomenon occurs. Several researchers have published papers on nonlinear coefficients of lead zirconate titanate (PZT). Jiang and Cao estimated a 3rd-order nonlinear coefficient (the coefficient of the 2nd-higher term) from the amplitude of 2nd harmonics generated in the sample when an ultrasonic wave of 10 MHz was transmitted into the bar sample from its end surface.4

Cho and Matsuno incorporated a PZT ceramic vibrator into a self-oscillation circuit. They estimated nonlinear coefficients by measuring the change of reactance and the resonance frequency of the self-oscillation circuit when electric fields and mechanical stress of 50 Hz were applied to the sample. However, all nonlinear coefficients up to the 4th order are imperfectly obtained.5 Beige and Schmidt estimated nonlinear coefficients when mechanical stress and electric fields were applied. However, they have not succeeded in separating their coefficients into 3rd- and 4th-order coefficients (the coefficients of 2nd- and 3rd-higher terms) when both coefficients are present simultaneously.6 Kugel and Cross tried to obtain the nonlinear coefficients related to the electric field. In order to prevent the influence of mechanical stress, they used a soft PZT material and applied electric fields having a frequency of 50 Hz which is much lower than the resonance frequency.7 The Reyleigh law is known to represent nonlinearity in magnetic materials. Damjanovic applied the Reyleigh law to the nonlinearities of permittivity and the piezoelectric constant which appeared when mechanical stress and electric field of low frequency were applied to a PZT sample.8 In those studies,5–6 since the frequencies of applied stress and electric field were less than 100 Hz, it is difficult to apply those results directly to piezoelectric ceramics driven at the resonance frequency.

Although efforts on the measurement of nonlinear coefficients have continued, as mentioned above, it is difficult to quantitatively estimate typical nonlinear phenomena, such as current jumping, using those nonlinear coefficients. Furthermore, although the relationship between voltage and current, which is obtained by solving the nonlinear piezoelectric equation, is essential for quantitative analysis of nonlinear phenomena, we cannot find any reports on it. This is probably due to the difficulty of solving the nonlinear piezoelectric equation.

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. Umeda et al. measured the change of linear constants in high-power vibration by the electrical transient response method.9–10 Takahashi and co-workers measured the change of linear constants depending on vibration velocity, and concluded that the nonlinear phenomena were caused by the change of elastic constants.11–12 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 have analyzed the current jump phenomenon and other nonlinear phenomena using an effective nonlinear coefficient that is constant for each piezoelectric vibrator.13 However, since the effective nonlinear coefficient depends on the vibrator size and the electrode surface even...
when the material composition is the same, it is impossible to use the effective nonlinear coefficient as a material constant. In the present study, we analyze the relationship between the nonlinear voltage between a pair of electrodes and a nonlinear coefficient, using the electrical equivalent circuit, in a piezoelectric rectangular bar. A nonlinear coefficient can be successively derived as a material constant in this analysis.

2. Nonlinear piezoelectric equation

There are two driving methods, constant-voltage driving and constant-current driving, in high-power driving at around the resonance frequency. Current jumping was observed in constant-voltage driving, and nonlinear voltages between pairs of electrodes were observed in constant-current driving, as typical nonlinear phenomena. Both phenomena are considered to originate from the same nonlinearity. Since current jumping is an unstable phenomenon, stable measurement is difficult in constant-voltage driving. On the other hand, nonlinear voltage between a pair of electrodes is measured when the sample is driven by a constant-current with sinusoidal waveform. Since the nonlinear voltages can be stably measured, constant-current driving is superior to constant-voltage driving for measurements in estimating the nonlinear coefficient.

There are four forms, d-form, g-form, e-form and h-form, in the piezoelectric equation. It is convenient to select electric flux density, \( D \), as an independent variable and electric field, \( E \), as a dependent variable in the measurement of sample voltage in constant-current driving. The combination of independent variable \( D \) and dependent variable \( E \) is satisfied in the g- and h-forms. Another independent variable in each of these two forms is stress, \( T \), in the g-form and strain, \( S \), in the h-form. Comparing \( T \) and \( S \), strain can be experimentally estimated by measuring the displacement of the end surface, whereas stress in the sample is difficult to estimate experimentally. Hence, the h-form which includes \( S \) as another variable was adopted.

We introduce nonlinear piezoelectric equations of the h-form by expanding Helmholtz’s free energy, \( A \), until the 4th-order term of strain, \( S \), and polarization, \( P \):

\[
E_3 = \left( \frac{\partial A}{\partial S} \right) P_1 + \left( \frac{\partial A}{\partial S} \right) S_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1^2
+ \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^3 + \frac{1}{6} \left( \frac{\partial A}{\partial S} \right) S_1^4 P_1
+ \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1^2 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1^2
+ \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1
+ \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1
+ \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1
+ \frac{1}{6} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1
+ \frac{1}{6} \left( \frac{\partial A}{\partial S} \right) S_1^3 P_1 + \frac{1}{2} \left( \frac{\partial A}{\partial S} \right) S_1^2 P_1
\]

where polarization can be approximated by electric flux density (\( P_i \approx D_i \)). Since \( S_1 \) is considered to be proportional to \( D_i \) at around the resonance frequency,\(^{13}\) all higher terms can be represented with only \( D_i \). Equations (1) and (2) are therefore rewritten as Eqs. (3) and (4), respectively.

\[
E_3 = -hS + \beta^3 D_1 + \gamma_0 D_1^2 + \xi_0 D_1^3 \quad (3)
\]

\[
T_1 = c^0 S_1 - hD_1 + \chi_0 D_1^2 + \delta_0 D_1^3 \quad (4)
\]

Here, \( h, \beta, \) and \( c^0 \) are linear coefficients: the piezoelectric \( h \)-constant, inverse permittivity and elastic stiffness, respectively. While \( \gamma_0, \xi_0, \chi_0, \) and \( \delta_0 \) are nonlinear coefficients in the representation using only \( D_i \), and they include the nonlinearities due to \( S_i \).

Since it is difficult to measure \( T_1 \) in samples, Eq. (4) is unsuitable for the estimation of nonlinearities. On the other hand, nonlinear coefficients can be determined by only electrical measurements in Eq. (3). Consequently, Eq. (3) is suitable for the estimation of nonlinear coefficients. Since Eqs. (3) and (4) are nonlinear piezoelectric equations, it is difficult to obtain the relationship between voltage and current by solving exactly them. Equations (3) and (4) were 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.

For the rectangular piezoelectric vibrator shown in Fig. 1 driven by a sinusoidal constant current having the resonance frequency of length-extensional 1/2\( \lambda \)-mode vibration, the boundary conditions are as follows: \( E_1 = E_2 = 0 \) and \( E_3 \neq 0 \) for electric fields, \( D_1 = D_2 = 0 \) and \( D_3 \neq 0 \) for electric flux density, \( T_1 \neq 0 \) and \( T_2 = T_3 = 0 \) for stress, \( S_1 \neq 0, S_2 \neq 0 \) and \( S_3 \neq 0 \) for strain. Since these boundary conditions are satisfied by the g-form piezoelectric equation, we transformed the g-form equation into the h-form equation. Equations (3) and (4) were rewritten as Eqs. (5) and (6) after the transformation.

\[
E_3 = -\frac{s_{31}}{s_{11}} S_1 + \beta^3 D_1 + \gamma_0 D_1^2 + \xi_0 D_1^3 \quad (5)
\]

\[
T_1 = -\frac{1}{s_{11}^2} S_1 - \frac{s_{31}}{s_{11}} D_1 + \chi_0 D_1^2 + \xi_0 D_1^3 \quad (6)
\]

Fig. 1. Configuration of the rectangular bar vibrator (\( l = 47.5 \text{ mm}, w = 6.9 \text{ mm}, b = 0.9 \text{ mm} \)).
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 $\gamma_{31}$ and $\zeta_{31}$ are effective nonlinear coefficients for the vibrator. The effective nonlinear coefficients are constant 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. (7).

\[
v_3 = \left( \omega L_1 - \frac{1}{\omega C_1} \right) I_3 \cos \omega t + R_1 I_3 \sin \omega t + \frac{\gamma_{31}}{\omega^3} I_3^3 \cos^2 \omega t - \frac{\zeta_{31}}{\omega^3} I_3^3 \cos^3 \omega t
\]  

(8)

Expanding $\cos^2 \omega t$ and $\cos^3 \omega t$ using trigonometric formulae for double and triple angles, Eq. (8) is rewritten as

\[
v_3 = \frac{\gamma_{31}}{2\omega^2} I_3^3 + \left( \omega L_1 - \frac{1}{\omega C_1} \right) I_3 \cos \omega t + R_1 I_3 \sin \omega t - \frac{3\zeta_{31}}{4\omega^3} I_3^3 \cos \omega t + \frac{\gamma_{31}}{2\omega^2} I_3^3 \cos 2\omega t - \frac{\zeta_{31}}{4\omega^3} I_3^3 \cos 3\omega t
\]  

(9)

A dc component and $2\omega$ component due to $\gamma_{31}$, and a $\omega$ component and $3\omega$ component due to $\zeta_{31}$, are generated as nonlinear voltages, respectively. We have quantitatively expressed nonlinear phenomena using Eq. (9).1)-3) In the case of length-extensional 1/2\(\alpha\)-mode vibration of the sample with whole-surface electrodes, as shown in Fig. 1, typical nonlinear phenomena, e.g., current jumping and the shift of resonance frequency, are due to $\zeta_{31}$ generating the $\omega$ component of voltage. Consequently, we have investigated $\zeta_{31}$ which is a coefficient of the 3rd higher term. The purpose of this work is to clarify the quantitative relationship between $\zeta_{31}$ used as a vibrator constant and $\zeta_{31}$ used as a material constant.

3. Experimental procedures

The composition of the prepared samples is 0.95Pb(\(\mathrm{Zr}_{0.51}\mathrm{Ti}_{0.49})\mathrm{O}_3-0.05\mathrm{Pb}(\mathrm{Sb}_{0.5}\mathrm{Nb}_{0.5})\mathrm{O}_3+0.015\mathrm{MnO}$. The samples were prepared using oxide powders as starting materials, by means of a conventional method, namely, mixing by wet ball-milling, calcining at 850°C for 2 h in air, grinding by wet ball-milling, pressing at 100 MPa, and firing at 1240°C for 2 h in air. After firing silver electrodes onto the upper and lower surfaces of the rectangular bars, the samples were poled under an electric field of 3 kV/mm for 10 min in a silicon oil bath kept at 120°C. Figure 1 shows the configuration of the rectangular bar vibrator used in this experiment. The motional impedance parameters ($L_1$, $C_1$, $R_1$) in the length-extensional 1/2\(\alpha\)-mode vibration, damped capacitance, $C_2$, mechanical quality factor, $Q_m$, electromechanical coupling factor, $k_{31}$, piezoelectric $d_{31}$ and $h_{31}$ constants, fundamental resonance and antiresonance frequencies, $f_r$ and $f_a$, elastic compliance, $s_{11}$, and relative permittivity, $\varepsilon_{33}/\varepsilon_0$, are listed in Table 1. These material constants were derived from the resonance-antiresonance characteristic measured at a low current level of less than 1 mA using an LF impedance analyzer (Hewlett-Packard, 4192A).

It is necessary to clarify where the nonlinear voltage source should be inserted in the equivalent circuit, to enable analysis using the equivalent circuit. In order to clarify the position of the nonlinear voltage source, the 3rd harmonic voltage, which is one of the nonlinear voltages, 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 3rd-harmonic voltage with a magnitude of 0.1 to 0.4% of the fundamental voltage; driving current consequently imposes a distortion via the 3rd-harmonic voltage. In order to prevent the application of the 3rd-harmonic voltage to the sample, a parallel resonance 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 three times the driving frequency. Since the parallel resonance circuit has an impedance above 100 kΩ at the frequency of the 3rd-harmonic voltage of the power amplifier (equal to the frequency of the 3rd-harmonic voltage generated in the sample), the parallel resonance circuit prevents the application of the 3rd-harmonic voltage generated in the power amplifier to the sample, and simultaneously enables us to take the 3rd-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 10 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\(\alpha\)-mode of the sample, and the signal of the function generator was amplified by a power amplifier (NF Electronics Instruments, 4020). A spectrum analyzer (Hewlett-Packard 4192A) was used for monitoring the output signal of the power amplifier.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Storage Scope DS-8807</th>
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<tbody>
<tr>
<td></td>
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<tr>
<td>Spectrum Analyzer HP ESA-L1600A</td>
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<tr>
<td>CH1</td>
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<tr>
<td>CH2</td>
<td></td>
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<tr>
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<tr>
<td>Lx</td>
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<tr>
<td>Cx</td>
<td></td>
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<tr>
<td>Power Amp. NF-4020</td>
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<td>Rb</td>
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</tbody>
</table>

Table 1. Piezoelectric Constants of the Sample

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<table>
<thead>
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<tbody>
<tr>
<td>$L_1$</td>
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<td>pF</td>
</tr>
<tr>
<td>$R_1$</td>
<td>Ω</td>
</tr>
<tr>
<td>$Q_m$</td>
<td></td>
</tr>
<tr>
<td>$k_{31}$</td>
<td>0.366</td>
</tr>
<tr>
<td>$f_r$</td>
<td>kHz</td>
</tr>
<tr>
<td>$f_a$</td>
<td>kHz</td>
</tr>
<tr>
<td>$d_{31}$</td>
<td>×10^-12 m/V</td>
</tr>
<tr>
<td>$h_{31}$</td>
<td>×10^4 N/C</td>
</tr>
<tr>
<td>$\varepsilon_{33}$</td>
<td>11.4</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>1270</td>
</tr>
</tbody>
</table>
Packard, ESA–L1500A) was used for the measurement of the magnitudes of fundamental and 3rd-harmonic voltages between electrodes of the sample.

4. Results and discussion

When a constant current of 29 mA$_{app}$ having the resonance frequency ($f_r$=35 kHz) drove two vibrators in the configuration shown in Fig. 1, the distorted voltage waves shown in Figs. 3(a) and (b) appeared between a pair of electrodes of the two samples. These distortions are due to the nonlinear voltages of $2\omega$ and $3\omega$ occurring in the samples due to $\delta_{D31}$ and $\delta_{D33}$. In particular, the component of $3\omega$ is marked in Fig. 3(b). Although the configurations of the two samples are almost the same, the magnitude of nonlinear voltages is different between the two samples.

In a rectangular piezoelectric vibrator, harmonic resonance appears at around the frequencies that are odd multiples of the fundamental resonance frequency, with the fundamental resonance. Since the frequency of 3rd-harmonic voltage is equal to 3 times the driving frequency, the magnitude of 3rd-harmonic voltage appearing between a pair of electrodes is probably affected by 3rd-harmonic resonance. The 3rd-harmonic resonance frequency is influenced by sample shape, being slightly different from 3 times the fundamental resonance frequency, $f_r$.

A schematic 3rd-harmonic resonance curve of Fig. 4(a) is representative, for convenience, of the curves of seven samples differing slightly in the relation between $3 \times f_r$ and $f_{3a}$, although the 3rd-harmonic resonance curves against frequency are slightly different among the seven samples. $f_{3a}$ is the 3rd-harmonic antiresonance frequency. These differences were caused by the slight change of the size and configuration of samples. When the seven samples with the configuration shown in Fig. 1 are driven at the fundamental resonance frequency by the constant-current method, the impedance at 3 times the driving frequency shows various values on the 3rd-harmonic resonance curve, as shown in Fig. 4(a). The plotted circles in Fig. 4(a) show where the frequency is 3 times the fundamental resonance frequency. Figure 4(b) shows the magnitude of 3rd-harmonic voltages, $V_{3ap}$, between a pair of electrodes when these seven samples were driven at the fundamental resonance frequency by the constant current of 29 mA$_{app}$. When $3 \times f_r$ equalled $f_{3a}$, the maximum $V_{3ap}$ was measured.

The electric equivalent circuit of a piezoelectric ceramic can be composed of motional impedance parameters originating in fundamental and 3rd-harmonic resonances ($L_1$, $C_1$, $R_1$, $L_3$, $C_3$, $R_3$) and damping capacitance, $C_s$. The results shown in Figs. 4(a) and 4(b) indicate where the nonlinear voltage source originating in nonlinear piezoelectricity should be inserted in the equivalent circuit. Figure 5 shows the equivalent circuit with the insertion positions (A, B, C) of the nonlinear voltage source $\delta_{D33}$.

Since the parallel resonance circuit having an impedance above 100 k$\Omega$ at the frequency of $3 \times f_r$ is connected to the sample, if the sample impedance is sufficiently lower than 100 k$\Omega$, the 3rd-harmonic voltage generated in the sample appears mostly between electrodes.

When $3 \times f_r$ equals the 3rd-harmonic resonance frequency, $f_{3r}$, the resonance resistance, $R_3$, of 3rd-harmonic resonance is very low compared with the impedance due to the damping capacitance, $C_s$. In this case, if the nonlinear

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**Fig. 3.** Voltage waveforms of the two samples driven by a constant current of 29 mA$_{app}$ with the resonance frequency $f_r$. The magnitude of 3rd-harmonic voltage is different between two samples even though they have almost the same configuration as shown in Fig. 1.

**Fig. 4.** Impedance values of seven samples at $3 \times f_r$ on the 3rd-harmonic resonance curve which is representative of the curves of seven samples for convenience (a), and the magnitudes of 3rd-harmonic voltage between electrodes of the seven samples (b). The 3rd-harmonic resonance and 3rd-harmonic antiresonance frequencies, $f_{3r}$ and $f_{3a}$, are about 103.9 and 104.5 kHz, respectively.
voltage source, $v_{31}$, is inserted at position (B), the 3rd-harmonic voltage is mostly applied to the damping capacitance and the minimum value must appear for $V_{3\text{ap}}$ in the seven samples. However, the minimum $V_{3\text{ap}}$ is not observed, as shown in Fig. 4(b). Hence position (B) is invalid.

In the case of position (C), when $3f_r$ equals the 3rd-harmonic antiresonance frequency, $f_{3a}$, the minimum value must appear for $V_{3\text{ap}}$. However, $V_{3\text{ap}}$ instead shows the maximum value, as shown in Fig. 4(b). Hence, position (C) is also invalid.

The results in Fig. 4(b) can be satisfactorily explained by inserting the nonlinear voltage source, $v_{31}$, at position (A). The magnitude of 3rd-harmonic voltages appearing between electrodes, $V_{3\text{ap}}$, can be expressed as Eq. (10) by considering the equivalent circuit and the magnitude of the 3rd component in Eq. (9).

$$V_{3\text{ap}} = \frac{\xi_{301}I_3}{Z_{CSS} + Z_{m3}}$$  

Here $Z_{CSS}$ and $Z_{m3}$ are the impedance of damping capacitance, $C_s$, and the 3rd motional impedance at $3\omega_r$. Since $Z_{CSS}$ and $Z_{m3}$ can be expressed as Eqs. (11) and (12), Eq. (10) is rewritten as Eq. (13).

$$Z_{CSS} = \frac{1}{j3\omega_r C_s}$$  

$$Z_{m3} = R_3 + j \left( 3\omega_r L_3 - \frac{1}{3\omega_r C_3} \right)$$  

$$V_{3\text{ap}} = -\frac{\xi_{301}I_3}{4\omega_r^3} \times \frac{1}{1 - 3\omega_r C_s \left( 3\omega_r L_3 - \frac{1}{3\omega_r C_3} \right) + j3\omega_r C_s R_3}$$

Equation (13) shows that the $V_{3\text{ap}}$ depends greatly on where the frequency of $3\omega_r$ is on the 3rd-harmonic resonance curve. When $3\omega_r$ equals $\omega_{3a} = 2\pi f_{3a}$, it is expressed as

$$3\omega_r C_s \left( 3\omega_r L_3 - \frac{1}{3\omega_r C_3} \right) = 1$$

Therefore, $V_{3\text{ap}}$ shows the maximum value. The curve fitted to the results of Fig. 4(b) by substituting an adequate value of $3.87 \times 10^{10} \text{ V/C}^2$ into $\xi_{301}$ in Eq. (13) is shown as a solid line in Fig. 4(b). The fitted curve agrees well with the measured values. Thus, by putting the nonlinear voltage source at position (A), the change of $V_{3\text{ap}}$ can be quantitatively explained.

The insertion position of $v_{31}$ suggests that $v_{31}$ is due to the current flowing through the motional admittance of fundamental resonance. This agrees with an argument in which the generation of 3rd-harmonic voltage in a piezoelectric transformer originates from the current flowing through the motional admittance. 10)

5. Theoretical analysis

As already mentioned, the purpose of this work is to obtain the quantitative relationship between $\xi_{301}$ and $\xi_{31}$. 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, $\omega_r$, by the constant-current method. Expressing $x$-distance, where the origin is at the center of the sample, $D_3$ is expressed

$$D_3 = -D_3 \cos \frac{\pi}{l} x \cdot \cos \omega_3 t$$

The nonlinear voltage generated in the sample due to the 3rd-highest term is expressed as

$$v_{31} = -b \xi_{303} D_3 = -b \xi_{303} D_3 \cos \frac{\pi}{l} \cdot x \cdot \cos^3 \omega_3 t$$

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

$$i_3 = \int_{-l/2}^{l/2} j_3 \, dx = \frac{2b \xi_{303} D_3}{\pi} \sin \omega_3 t = i_3 \sin \omega_3 t$$

From Eq. (17), we obtain

$$D_3 = -\frac{i_3 \pi}{2b \xi_{303}}$$

Substituting Eq. (18) into Eq. (16), we get

$$v_{31} = -b \xi_{303} D_3 \cos \frac{\pi}{l} \cdot x \cdot \cos^2 \omega_3 t$$

$$= -b \xi_{303} \frac{I_3^3}{8\pi^3 \omega_3^3 \cos \omega_3 t} \left\{ \frac{9}{16} \cos \frac{\pi}{l} \cdot x \cdot \cos \omega_3 t + \frac{3}{16} \cos \frac{3\pi}{l} \cdot x \cdot \cos 3\omega_3 t + \frac{1}{16} \cos \frac{3\pi}{l} \cdot x \cdot \cos 3\omega_3 t \right\}$$

Thus, two nonlinear voltages having two frequency components, $\omega_3$ and $3\omega_3$, exist in $v_{31}$ due to the 3rd-highest term, and they have cosine distributions in the length direction.

It is necessary to investigate how voltage is measured between electrodes when nonlinear voltages with cosine distribution exist in the sample having an electrode on each of the upper and lower surfaces. Since each angular frequency, $\omega_r$ and $3\omega_r$, is under the influence of each piezoelectric resonance, the distribution of motional impedance consequently exists in the sample. Since the sample is resonant at $\omega_r$, there is no reactance and only $R_3$ exists against the fundamental voltage with $\omega_r$. Since $R_3$ exhibits a cosine distribution, the resistance per meter in the length direction is represented by $R_{1(x)}$ which is expressed as

$$R_{1(x)} = R_{1(0)} \frac{1}{\cos \frac{\pi}{l} x}$$

where $R_{1(0)}$ is the resistance per meter at $x=0$. On the other
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hand, the impedance against the 3rd-harmonic voltage has reactance components since the 3rd-harmonic resonance angular frequency, \( \omega_3 \), is different from \( 3\omega_1 \). The impedance per meter in the length direction, \( Z_{m3}(x) \), is expressed as

\[
Z_{m3}(x) = Z_{m3(0)} \frac{1}{\cos \frac{3\pi}{l} x}
\]

where \( Z_{m3(0)} \) is the impedance per meter at \( x=0 \).

In the sample having distributions of both nonlinear voltage and impedance, the voltage measured between electrodes can be calculated using Millman’s theorem.\(^{15}\) By considering the insertion position of the nonlinear voltage source and the impedance distribution in the sample, the equivalent circuit, which is necessary for determining the voltage between electrodes (\( v_{h11} \)) due to \( v_{h11} \) of the first term in Eq. (19), is shown in Fig. 6. The magnitude of \( v_{h11} \) is expressed as in Millman’s theorem:

\[
v_{h11} = v_{h11} \cdot \cos \omega \cdot \alpha \cdot t
\]

Since both distributions are symmetric on both sides of \( x=0 \), the calculation was performed in the range of \( x=0 \) to \( x=1/2 \). Furthermore, since \( R_1 \) is too small for comparison with \( 1/\omega_1 C_1 \), the influence of the impedance due to the damping capacitance, \( Z_{cs} = 1/\omega_1 C_1 \), can be neglected. By including the resistance distribution of Eq. (20) and the nonlinear fundamental voltage distribution of the first term of Eq. (19) in Millman’s theorem, \( V_{h11} \) is obtained as follows:

\[
V_{h11} = \frac{V_{h11}(0) R_{1(0)}}{R_{1(0)} + \frac{1}{1} R_{1(1)} + \frac{1}{2} R_{1(2)} + \cdots + \frac{1}{n} R_{1(n)}} + \frac{V_{h11}(1) R_{1(1)}}{R_{1(1)} + \frac{1}{1} R_{1(1)} + \frac{1}{2} R_{1(2)} + \cdots + \frac{1}{n} R_{1(n)}} + \cdots + \frac{V_{h11}(n) R_{1(n)}}{R_{1(n)} + \frac{1}{1} R_{1(1)} + \frac{1}{2} R_{1(2)} + \cdots + \frac{1}{n} R_{1(n)}}
\]

In the angular frequency component of \( \omega_3 \), since \( R_1 \) is too small for comparison with \( 1/\omega_3 C_3 \), the influence of the impedance due to the damping capacitance, \( Z_{cs} = 1/\omega_3 C_3 \), can be neglected. However, in the case of disagreement between \( 3\omega_1 \) and the 3rd-harmonic resonant angular frequency, \( \omega_3 \), since the motional impedance of the 3rd-harmonic resonance is large, the influence of the impedance due to the damping capacitance at \( 3\omega_1 \), \( Z_{cs} = 1/\omega_3 C_3 \), should be carefully considered. The magnitude of \( v_{h13} \) between electrodes, due to \( v_{h13} \) of the third term in Eq. (19), is expressed as follows:

\[
V_{h13} = \frac{3}{4} \pi \cdot \frac{Z_{cs}}{Z_{cs} + Z_{m3}}
\]
Since $V_{h11}(0)$ and $V_{h33}(0)$ are the magnitudes of $v_{h11}$ and $v_{h33}$ at $x=0$, they are expressed from Eq. (19) as follows.

\begin{equation}
V_{h11}(0) = \frac{9}{16} b_{011} \frac{L^3}{8f_w \omega_0^3} \tag{28}
\end{equation}

\begin{equation}
V_{h33}(0) = \frac{1}{16} b_{031} \frac{L^3}{8f_w \omega_0^3} \tag{29}
\end{equation}

Substituting Eqs. (28) and (29) into Eq. (27), $v_{\xi_{ap}}$ is expressed as follows.

\begin{equation}
v_{\xi_{ap}} = \frac{3}{512} b_{011} \frac{L^3}{f_w^3 \omega_0^3} \left\{ 3 \cos \omega_0 t + \frac{Z_{CS}}{Z_{CS} + Z_{3m}} \cos 3\omega_0 t \right\} \tag{30}
\end{equation}

$V_{h1ap}$ and $V_{h3ap}$ are the magnitudes of the fundamental and 3rd-harmonic voltages between a pair of electrodes.

On the other hand, we have explained nonlinear phenomena of a piezoelectric vibrator in terms of the effective 3rd-nonlinear coefficient, $\xi_{D31}$, in Eqs. (8) and (9). When the sample was driven at $\omega = \omega_0$, the nonlinear voltages, $v_{\xi_{ap}}$, between electrodes due to $\xi_{CS}$ in Eq. (8) are expressed as Eq. (31), by considering the influence of the 3rd-motional impedance, $Z_{3m}$, and the impedance of the damping capacitance, $Z_{3c}$, in Eq. (10).

\begin{equation}
v_{\xi_{ap}} = -\frac{3}{4} b_{011} \frac{L^3}{f_w^3 \omega_0^3} \left\{ \frac{Z_{CS}}{Z_{CS} + Z_{3m}} \cos 3\omega_0 t \right\} \tag{31}
\end{equation}

Comparing Eq. (31) with Eq. (30), the sign of the second term is different. This disagreement is due to the disregard of the phase relation between fundamental and 3rd-harmonic voltages occurring in the process of deriving Eq. (31). When regarding the phase relation, Eq. (30) is correct. However, there is no problem in the measurement which we have continued to obtain the value of $\xi_{D31}$, since only the magnitudes of fundamental and 3rd-harmonic voltages are necessary in the measurement. From Eqs. (30) and (31), the relationship between $\xi_{D31}$ and $\xi_{D31}$ is obtained as

\begin{equation}
\xi_{D31} = -\frac{3}{128} b_{011} \frac{L^3}{f_w^3} \tag{32}
\end{equation}

There are 3 methods of obtaining the values of $\xi_{D31}$ and $\xi_{D31}$: (1) measure the magnitudes of fundamental voltages, $V_{h1ap}$, due to $\xi_{D31}$ between electrodes, (2) measure the magnitudes of 3rd-harmonic voltages, $V_{h3ap}$, between electrodes, (3) measure the fundamental resonance frequency change due to the occurrence of $V_{h1ap}$. We will discuss the comparison of these 3 methods in other papers.

As shown in Eqs. (3) and (5), $\xi_{D31}$ is the coefficient when higher terms are represented with only $D_3$ under the condition that $S_1$ is proportional to $D_3$. The representation of all higher terms with only $S_1$ under the condition that $S_1$ is proportional to $D_3$ is expressed as

\begin{equation}
E_3 = -\frac{g_{31}}{S_{11}} S_1 + \beta_{31} D_3 + \gamma_{S3} S_1^3 + \xi_{S31} S_1^3 \tag{33}
\end{equation}

where $\gamma_{S3}$ and $\xi_{S31}$ are the coefficients in the representation with only $S_1$ for all higher terms. The relationship between $S_1$ and $D_3$ at the resonance frequency is expressed as Eq. (34) by neglecting higher terms of Eq. (33) and substituting $E_3 = 0$.

\begin{equation}
S_1 = \frac{g_{31} D_3^{3/2}}{\gamma_{S3}} \tag{34}
\end{equation}

Substituting Eq. (34) into Eq. (33), and comparing the obtained equation with Eq. (5), the relationship between $\gamma_{S3}$ and $\gamma_{D31}$, and that between $\xi_{S31}$ and $\xi_{D31}$, can be expressed as

\begin{equation}
\gamma_{S3} = \left( \frac{g_{31}}{S_{11}} \right)^{3/2} \gamma_{D31} \tag{35}
\end{equation}

\begin{equation}
\xi_{S31} = \left( \frac{g_{31}}{S_{11}^{3/2}} \right)^{3} \xi_{D31} \tag{36}
\end{equation}

In order to estimate $\gamma_{D31}$, it is necessary to measure 2nd-harmonic voltage with the $2\omega_0$ component, as shown in Eq. (9). However, it is difficult to extract the 2nd-harmonic voltage from the sample with whole surface electrodes, since it has no resonance at around $2\omega_0$. Furthermore, it is also difficult to derive $\gamma_{S3}$ from the measured 2nd-harmonic voltage since it is difficult to estimate the internal impedance of the sample relative to the 2nd-harmonic voltage. Half-surface electrodes are essential for the resonance to appear at around $2\omega_0$. By measuring the 2nd-harmonic voltage in the sample with half-surface electrodes, $\gamma_{D31}$ can be obtained by an analysis similar to that described in this paper.

6. Conclusions

When a piezoelectric rectangular bar was driven at the resonance frequency by the constant-current method, nonlinear fundamental and nonlinear 3rd-harmonic voltages between electrodes can be derived as a material constant by using the magnitudes of the nonlinear voltages between electrodes in Eq. (30).

References