Simulation of current-jumping phenomena in PbTiO₃–Pb(Sc₁/₂Nb₁/₂)O₃ system piezoelectric ceramics

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1. Introduction

The suppression of the appearance of the nonlinear phenomena is a critical issue for the practical uses of the piezoelectric ceramic devices driven at a high power such as piezoelectric transformers and ultrasonic motors. The current-jumping phenomena, the generation of higher harmonic voltages, the changes of observed material constants, and the rapid temperature rising of the devices are typical nonlinear phenomena. These phenomena are the main causes of the unstable operation and the increase of energy loss in high-power vibrations. In order to improve the reliability and the driving efficiency of the devices, not only the search for piezoelectric materials which have a smaller nonlinear piezoelectricity, but also the exact prediction of the driving conditions inducing the nonlinear phenomena is indispensable. The prediction is obtained from the quantitative estimation of the nonlinearity.

We have quantitatively estimated the nonlinearity by defining a new nonlinear piezoelectric coefficient of third-higer term for the electrical field, \( \xi_{31} \), as a material constant. This coefficient quantifies the nonlinear movement of non-180° domain walls. Unlike the analysis focused on the change of linear piezoelectric constants by Umeda and Takahashi, our analysis using \( \xi_{31} \) enables us to predict the driving condition inducing the nonlinear phenomena not only to compare the quantitative nonlinearity among the materials. We have proposed the calculating equations introduced from \( \xi_{31} \) to simulate the current-jumping phenomena and the change of resonance frequency.

However, it has been found that the values calculated using \( \xi_{31} \) do not exactly correspond to the current-jumping phenomena measured in some piezoelectric ceramics. This fact suggests that another nonlinear piezoelectricity exists not only within the previous nonlinear piezoelectricity estimated with \( \xi_{31} \). Hence, a new theoretical analysis was performed in this study, in which another nonlinear piezoelectricity estimated with a new nonlinear coefficient of third-higer term for mechanical stress, \( \xi_{31} \), was added to the previous one estimated with \( \xi_{31} \). Furthermore, the loss term due to the nonlinearity was quantitatively expressed by using \( \xi_{31} \) as a complex number. By using the imaginary part of \( \xi_{31} \), the drop point of the current was exactly simulated in current-jumping phenomena.

The \( \xi_{31} \) values have a negative correlation with the linear values in piezoelectric ceramics. Since PbTiO₃–Pb(Sc₁/₂Nb₁/₂)O₃ (PT–PSN) ceramics are reported to be ferroelectric relaxers having a large \( \xi_{31} \), its nonlinear piezoelectricity is expected to be small. In this study, the theoretical analysis mentioned above was performed by using experimental data for PT–PSN ceramics.

2. Previous theory of current-jumping and its problems

We have analyzed the nonlinear piezoelectricity using the 3rd nonlinear piezoelectric equation transformed into the \( h \)-form equation from the \( g \)-form equation, which is expressed as Eq. (1).

\[
E_3 = -\frac{S_{11}}{s_{11}} S_1 + \beta_3 S_3 D_3 + \gamma_{33} D_3^2 + \xi_{31} D_3^3
\]

In Eq. (1), the \( \gamma_{33} \) and \( \xi_{31} \) are the intrinsic nonlinear coefficients of the material constants. The \( g_{33} \), \( \beta_3 \), and \( s_{11} \) are the piezoelectric \( g \) constant, inverse permittivity, and elastic compliance, respectively. The \( E_3 \) and \( D_3 \) are the electric field and electric flux density in the thickness direction, and \( S_1 \) is the mechanical strain in the transverse direction in the coordinate system as shown in Fig. 1(a). Since only the third nonlinear term in Eq. (1) induces typical nonlinear piezoelectric phenomena such as current-jumping phenomena and the change of resonance frequency, the second nonlinear term is negligible. When the sample is driven by a constant current at the fundamental resonance frequency, Eq. (2) can be obtained by superposing the nonlinear terms on the solution of linear terms, which is one of...
Here, $i_3$ is driven at the resonance frequency, and it is expressed using the effective nonlinear coefficient for the rectangular vibrator, and it is expressed using $\zeta_{31}$ as Eq. (3). \(^{11)\),\(^{12)}\),\(^{21)}\)

$$\zeta_{31} = \frac{nb}{A^2} \xi_{31}$$  

(3)

Here, $A = l \times a$ is the electrode area, and $l$, $a$, and $b$ are the sample length, width, and thickness, respectively. The $n$ is the conversion factor obtained by considering the electrode configuration, and is equal to $3\pi^2/128$ when the rectangular samples have the whole-surface electrodes. \(^{21)}\)

When a hard material of high $Q_a$ is driven at the resonance frequency, the sample current, $i_3$, is almost equal to the current through the motional impedance branch of the equivalent circuit of the piezoelectric ceramic, $i_3'$, as shown in Fig. 1(b). When the sample is driven by a current, $i_3 = I_o \sin(\omega t)$, the fundamental frequency component of $v_3$ obtained from Eq. (2), $V_0 \sin(\omega t + \theta)$ is expressed as the following equations.

$$V_0 \sin(\omega t + \theta) = \left(\frac{\omega L_3 - 1}{\omega C_1} I_0 \cos(\omega t) + R_i I_0 \sin(\omega t) - \frac{3\zeta_{31}}{4\omega^3} I_0^3 \cos(\omega t)\right)$$

$$V_0 \sin(\omega t + \theta) = \sqrt{\left(\frac{\omega L_3 - 1}{\omega C_1} I_0 - \frac{3\zeta_{31}}{4\omega^3} I_0^3\right)^2 + (R_i I_0)^2} \sin(\omega t + \theta)$$

where, $\theta = \tan^{-1} \left(\frac{\omega L_3 - 1}{\omega C_1 \frac{3\zeta_{31}}{4\omega^3} I_0^2}{R_i}\right)$  

(4)

From Eq. (4), the relationship between the voltage amplitude and current amplitude is given as Eq. (5). \(^{14)}\)

$$V_0 = \sqrt{\left(\frac{\omega L_3 - 1}{\omega C_1} I_0 - \frac{3\zeta_{31}}{4\omega^3} I_0^3\right)^2 + (R_i I_0)^2}$$  

(5)

We have simulated the current-jumping phenomena using Eq. (5). \(^{14)}\)

The resonance frequency, $f_3$, at which $\theta$ in Eq. (4) becomes equal to zero, decreases with the increase of current, and the relationship between the resonance frequency and current amplitude is expressed as Eq. (6). \(^{11)}\)

$$\omega_3 = 2\pi f_3 = \sqrt{1 + \frac{1}{3L_3 C_3^2 \xi_{31} I_0^2} \frac{2L_3 C_3}{I_0}}$$  

(6)

Recently, it was found that the current-jumping simulation calculated with Eq. (5) and the resonance frequency simulation calculated with Eq. (6) do not agree with the experimental values in some materials. This indicates that the nonlinear piezoelectricity is imperfectly estimated in our previous calculation using only $\zeta_{31}$ value.

### 3. Experimental

The material compositions used in this study are $x$PbTiO$_3$–(1-)$x$PbSc$_{1/2}$Nb$_{1/2}$O$_3$. The MnO at amount of 1.5 mol% was added as an acceptor. The samples were fired at 1250°C for 2 h in air. The standard sample dimensions of a rectangular bar were 30 $\times$ 4 $\times$ 1 mm$^3$ as shown in Fig. 1(a). The standard aging time before the measurements after poling (3 kV/mm, 120°C, 10 min) was 48 h at room temperature. The crystal structures were determined from the powder X-ray diffraction (XRD) patterns of the sintered samples. The linear piezoelectric constant, $d_{31}$, the electromechanical factor, $k_{31}$, and mechanical quality factor, $Q_a$ were measured by the resonance-antiresonance method on the basis of IEEE standards using an LF-impedance analyzer (HP, 4192A) under a small signal.

In order to measure the $\zeta_{31}$ values, the sample was driven at the fundamental resonance angular frequency of the length-extensional 1/2 $l$ mode, $\omega_3$, by a signal generator (HP, 3325B) and a power amplifier (NF, 4020) in the constant-current circuit, as shown in Fig. 2(a). Here, $\omega_3$ is the resonance angular frequency when the samples were driven under a small signal. The waveforms of sample current and sample voltage were simultaneously accumulated into a digital storage scope (Lecroy, LT3444). \(^{14)}\) From the amplitude of the third harmonic voltage, $V_{3s}$, observed across the samples driven by the constant current of $\omega_3$, $\zeta_{31}$ was calculated using Eq. (7). Furthermore, $\zeta_{31}$ was converted to $\zeta_{31}'$ using Eq. (3).

$$V_{3s} = \frac{\xi_{31}}{4\omega_3^3} \frac{Z_{C_3}}{Z_{C_3} + Z_{m_3}}$$

where,

$$Z_{m_3} = R_3 + j \left(3L_3 \omega_3 - \frac{1}{3C_3 \omega_3}\right)$$

$$Z_{C_3} = \frac{1}{j3\omega_3 C_3}$$

(7)

Here, $L_3$, $C_3$, and $R_3$ are the motional impedance parameters for the 3rd resonance, and $Z_{m_3}$ and $Z_{C_3}$ are the motional impedance for the 3rd resonance and the impedance due to the damping capacitance at $3\omega_3$. A high-power driving induces the slight change of the elastic compliance in the samples. The $\zeta_{31}'$ was calculated using the equation in which the change of the elastic compliance was compensated. \(^{14)}\)
For the measurement of the current-jumping phenomena, the samples were driven at a constant voltage of sinusoidal waveform using the constant-current circuit, as shown in Fig. 2(b). After the amplification of the voltage measured across the resistance, \( R_a \), its signal was rectified with a full-wave rectifier circuit, and the dc signal was stored into LT–344. The sweep rate of the driving frequency is 1 kHz/s. All the measurements were done at room temperature.

4. Results and discussion

4.1. Nonlinear piezoelectricity of PZT–PSN

The XRD profiles of the sintered samples are shown in Fig. 3. All of the samples have a single phase of perovskite structure. Judging from the diffraction lines at around 45°, the crystal phase of the samples with the PT contents larger than 44 mol% is tetragonal, and that of the samples with the PT contents smaller than 42 mol% is rhombohedral. The sample having a PT content of 43 mol% shows a mixed phase of tetragonal and rhombohedral, being identified as the morphotropic phase boundary (MPB) composition.

Figure 4 shows the changes of \( Q_m \) and \( d_{31} \) as functions of PT content. When the PT/PSN ratio is 43/57, namely, the ratio of MPB composition, the maximum \( d_{31} \) and the minimum \( Q_m \) were observed. The dependence of \( \xi_{31} \) on the PT content ratio is illustrated in Fig. 5. The minimum \( \xi_{31} \) value observed in the sample of MPB composition as the same as in the previous cases of the PZT system ceramics.\(^{12,13,15}\) This is due to the minimum anisotropy of the crystal structure at the MPB composition. It has been demonstrated that the nonlinear piezoelectric phenomena are caused by the nonlinear movements of non-180° domain walls.\(^{12,13}\) When the crystal anisotropy is larger, it is commonly believed that the nonlinear piezoelectricity induced by the nonlinear movements of non-180° domain walls is also larger. The minimum \( \xi_{31} \) value observed in this study \((1.7 \times 10^{12} \text{ Vm}^5/\text{C}^3)\) was smaller than the minimum value previously measured in the PZT ceramics \((>2 \times 10^{12} \text{ Vm}^5/\text{C}^3)\).

Figure 6 represents the current-jumping phenomena in the samples of PT/PSN ratios of (43/55), (43/57), and (41/59) when they were driven using the constant-voltage circuit as shown in Fig. 2(b). The applied electric field is 4 V/mm. The broken curves in Fig. 6 were calculated with Eq. (5), which is our old equation for the simulation of the current-jumping phenomena.
Although the broken line agreed well with the measured values in (45/55) sample, the considerable discrepancy was observed between the broken line and the measured values in the (41/59) sample. Figure 7 illustrates the changes of resonance frequencies in these three samples driven using the constant-voltage circuit, broken and solid lines indicate the curves calculated using Eq. (5) and Eq. (23), respectively.

Figure 7 illustrates the changes of resonance frequencies in these three samples driven using the constant-current circuit as shown in Fig. 2(a). The curves calculated with Eq. (6) are drawn with broken lines. The $f_0$ indicates the resonance frequency at a time when the samples are driven at a small signal. The broken line agreed with the measured values (open circles) in (45/55) sample as well as in the case of the current-jumping phenomena of Fig. 6. On the other hand, the considerable discrepancy between the broken line and measured values appeared in the (41/59) sample in the large current region. These discrepancies between the broken line and the measured values are inconspicuous in the PZT samples having a relatively large $|\zeta_{D31}|$. These facts suggest that although its contribution to the nonlinear phenomena is relatively smaller compared with $\zeta_{D31}$, other nonlinear piezoelectricity exists which is expressed by another nonlinear piezoelectric coefficient except $\zeta_{D31}$.

In addition to Eq. (1) having the output of electrical field, the following equation having the output of mechanical stress exists in the third nonlinear piezoelectric equations transformed into the $h$-form equation from the $g$-form equation in rectangular-bar samples driven in $31$ mode.\(^5\)

$$T_1 = \frac{1}{s_{11}} S_1 - \frac{g_{11}}{s_{11}} D_3 + \frac{\chi_{D31}}{s_{11}} D_3^2 + \frac{\zeta_{D31}}{s_{11}} D_3^3$$ \hspace{1cm} (8)

Equation (8) has a nonlinear piezoelectric coefficient of third-higher term, $\zeta_{D31}$. The effects of $\zeta_{D31}$ on the current-jumping phenomena and the change of resonance frequency were investigated.
4.2. Theoretical analysis including the nonlinear coefficient of third-higher term for mechanical stress

When the rectangular piezoelectric vibrators being stress-free on both sides were driven at a sinusoidal electrical field, $E_1$, at the resonance frequency of length-extensional 1/2 $\lambda$-mode vibration in the transverse direction, the boundary conditions are as follows: $E_1 = E_2 = 0$, $E_3 \neq 0$, $T_1 \neq 0$, $T_2 = T_3 = T_4 = 0$, $S_1 \neq 0$, $S_2 = 0$, $S_3 = 0$. Equations (9) and (10) satisfying these boundary conditions are well-known as $d$-form equations.

\[
S_1 = d_{31}E_3 + s_{11}E_1 \quad \text{(9)}
\]
\[
D_1 = e_{13}E_3 + d_3T_1 \quad \text{(10)}
\]

Here, $s_{11}$ and $e_{13}$ are elastic compliances in the transverse direction and permittivity in the thickness direction, respectively.

As mentioned in Chapter 2, only the nonlinear coefficient of third-higher term induces the typical nonlinear piezoelectric phenomena. Adding the third nonlinear term of Eq. (8), $\zeta_{D3}D_3^3$, to $T_1$ obtained by solving Eq. (9), we obtain the following equation.

\[
T_1 = \frac{s_{11}}{s_{11}^b}E_1 + \frac{d_{31}}{s_{11}^b}E_3 + \zeta_{D3}D_3^3 \quad \text{(11)}
\]

By adding $\zeta_{D3}D_3^3$ to the linear solution for mechanical stress obtained from Eqs. (9) and (10), we aim to obtain the solution for $T_1$ of Eq. (11). The $\chi$ indicates the position in the length direction of the sample where the origin is at the center of the sample. When the electrical field, $E_1 = b \frac{V_0}{b} \cos \omega t / b$, is applied to the sample, the linear solution of $T_1$ around the resonance frequency is given with the acoustic velocity, $v$, as follows:

\[
T_1 = \frac{d_{31}V_0}{s_{11}^b \cos \frac{\omega}{2} l} \cos \left(\frac{\omega}{l} \chi\right) e^{i\omega t} - \frac{d_{31}V_0}{s_{11}^b \cos \frac{\omega}{2} l} e^{i\omega t} \quad \text{(12)}
\]

On the other hand, $D_3$ in the third-higher term of Eq. (11) is the electric flex density induced on the electrode surfaces. Since $D_3$ is distributed with cosine distribution in the length direction of the sample driven at the fundamental resonance frequency, it is expressed as follows:

\[
D_3 = D_0 \cos \left(\frac{\pi}{l} \chi\right) e^{i\omega t} \quad \text{(13)}
\]

Here, $\alpha$ is the phase difference between $V_1$ and $D_0$. Expressing the sample current in the vector, $\mathbf{I}_3$, it is given by

\[
\mathbf{I}_3 = a \int_0^1 \frac{d}{dt} x = \int_0^1 \frac{a}{\pi} \cos \left(\frac{\pi}{l} x\right) \frac{d}{dt} x = j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})} = j I_0 e^{i(\omega t + \frac{\pi}{2})} \quad \text{(14)}
\]

The relationship of the amplitudes between the electric flex density and the current is therefore expressed by Eq. (15).

\[
D_0 = \frac{\pi I_0}{2 a o l} \quad \text{(15)}
\]

After substituting Eqs. (12) and (13) into Eq. (11), extracting only the fundamental frequency component, the following equation is obtained.

\[
T_1 = \frac{d_{31}V_0}{s_{11}^b \cos \frac{\omega}{2} l} \cos \left(\frac{\omega}{l} \chi\right) e^{i\omega t} - \frac{d_{31}V_0}{s_{11}^b \cos \frac{\omega}{2} l} e^{i\omega t} + \zeta_{D3}D_3^3 \left[\frac{9}{16} \cos \left(\frac{\pi}{l} \chi\right) + \frac{3}{16} \cos \left(\frac{3\pi}{l} \chi\right)\right] e^{i(\omega t + \frac{\pi}{2})} \quad \text{(16)}
\]

Substituting Eq. (16) into Eq. (10), because of $e_{33} = e_{33}^T \frac{d_{31}^2}{s_{11}^E}$, we obtain Eq. (17).

\[
D_3 = \frac{e_{33}^T}{b} \frac{d_{31}^2}{s_{11}^E} V_0 e^{i\omega t} + \frac{d_{31}^2 V_0}{s_{11}^b \cos \frac{\omega}{2} l} \cos \left(\frac{\omega}{l} \chi\right) e^{i\omega t} + \frac{d_{31}^2 \zeta_{D3} D_3^3}{s_{11}^b \cos \frac{\omega}{2} l} \left[\frac{9}{16} \cos \left(\frac{\pi}{l} \chi\right) + \frac{3}{16} \cos \left(\frac{3\pi}{l} \chi\right)\right] e^{i(\omega t + \frac{\pi}{2})} \quad \text{(17)}
\]

From Eq. (17), $\mathbf{I}_3$ containing the contribution of $\zeta_{D3}$ is given by

\[
\mathbf{I}_3 = a \int_0^1 \frac{d}{dt} x = \int_0^1 \frac{a}{\pi} \cos \left(\frac{\pi}{l} x\right) \frac{d}{dt} x = j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})} + j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})} + j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})} = j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})} \quad \text{(18)}
\]

Because of, $D_0 = \frac{\pi I_0}{2 a o l}$, $\mathbf{I}_3 = j \frac{2 a o D_0}{\pi} e^{i(\omega t + \frac{\pi}{2})}$, $V_3 = V_0 e^{i\omega t}$ with the vector, $\mathbf{I}_3$, and transforming Eq. (19), we obtain Eq. (20).
The first and the second terms in the right side of Eq. (20) indicate the currents through the damping capacitance branch and motional impedance branch in the equivalent circuit of the piezoelectric vibrator shown in Fig. 1(b). We obtain accordingly the admittance, Y, as follows.

\[
Y = \frac{I}{V} = \frac{j}{1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \tan \left( \frac{\omega L_1}{2a} \right)} \times \left( C_0 \omega + \frac{2 \alpha \omega \tan \left( \frac{\omega L_1}{2a} \right)}{s_1^2 b} \right)
\]  

(20)

From Eq. (21), the motional impedance, \(Z_m\) is given by the following equation.

\[
Z_m = \frac{1}{Y_m} = \frac{1}{1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \tan \left( \frac{\omega L_1}{2a} \right)} \left( \alpha L_4 - \frac{1}{\omega C_1} \right)
\]  

(22)

The term of \(1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \tan \left( \frac{\omega L_1}{2a} \right)\) in Eq. (22) is the correction term due to \(\xi_{D31}\). Since \(\xi_{D31}\) and \(d_3\) are both negative values, \(L_4\) decreases and \(C_1\) increases when the current increases.

Rewriting Eq. (5) using the motional impedance expressed by Eq. (22), the relationship between the amplitude of voltage, \(V_0\), and that of current, \(I_0\), in the constant voltage driving, is given by

\[
V_0 = \left[ \left( 1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \frac{1}{\omega C_1} \right) \left( \alpha L_4 - \frac{1}{\omega C_1} \right) + \frac{3 \xi_{D31}^2 \omega^2 I_0^2}{4a^2} \right]^{1/2} \times (R I_0)^2.
\]  

(23)

The calculation for the current responses to frequency of the current-jumping phenomena was repeated by substituting various values into \(\xi_{D31}\) of Eq. (23). When the calculated curve became most closely to the measured curve, the value used in the calculation was adopted as the true value of \(\xi_{D31}\). The solid lines in Fig. 6 illustrate the curves calculated with the \(\xi_{D31}\) values obtained by this method. Compared with the broken lines, which do not include the contribution of \(\xi_{D31}\), the solid lines well agree with the measured curves.

By adding the contribution of \(\xi_{D31}\) to Eq. (6) for the simulation of the change of resonance frequency, \(f_1\), Eq. (24) is obtained.

\[
\omega_0 = 2 \pi f_1 = \sqrt{1 + \frac{1}{2L_2 C_1} \left[ 1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \right]^{1/2} \frac{3 \xi_{D31}^2 \omega^2 I_0^2}{4a^2} \times (R I_0)^2}
\]  

(24)

The solid curves in Fig. 7 were drawn by substituting the \(\xi_{D31}\) values into Eq. (24), which values gave the solid curves in Fig. 6. Although the considerable discrepancy between the measured curve and the curve calculated with Eq. (6) was observed in the (41/59) sample, the curve calculated with Eq. (24) corresponded exactly to the measured curve. The calculated curves for the simulations of the two different nonlinear phenomena, namely, the current-jumping and the change of resonance frequency, were simultaneously corrected with the same \(\xi_{D31}\) value, and agreed well with the measured curves, respectively. Hence, we concluded that the theoretical analysis based on \(\xi_{D31}\) was valid.

The PT-content dependence of \(\xi_{D31}\), which was obtained in the method mentioned above, is shown in Fig. 8. The value of \(\xi_{D31}\) increased in the rhombohedral side. This result indicates that the appearance of nonlinearity due to \(\xi_{D31}\) is related to \(71^\circ\) and \(109^\circ\) domain walls, which are non-180° domain walls, in the rhombohedral region. However, its mechanism of the non-180° domain walls is not clear yet.

4.3. Expression of loss term by complex \(\xi_{D31}\)

Although the solid curves in Fig. 6 reproduce well the configuration of the measured current responses to the frequency, the current values of the drop points are smaller than the current values predicted by the solid lines. This indicates that the practical loss component in the samples driven at a high power is larger than \(R_1\), which is the resistance resonance at a time when a driving voltage is sufficiently small to prevent the appearance of nonlinear phenomena. The increase of the loss component is probably caused by the phase delay of the nonlinear movements of non-180° domain walls, and this is also considered to induce the rapid temperature rising\(^{6,7}\) and the decrease of \(\xi_{D31}^{\prime\prime}\) of the devices driven at a high power. The phase delay is quantitatively expressed by using \(\xi_{D31}^{\prime\prime}\) as a complex number. Substituting the complex \(\xi_{D31}^{\prime\prime} = \xi_{D31}^{\prime\prime} - j \xi_{D31}^{\prime\prime}\) into Eq. (23), the relationship between \(V_0\) and \(I_0\) is given by Eq. (25).

\[
V_0 = \left[ \left( 1 - \frac{d_3^2 \omega \xi \pi^2}{8a^3 \omega^2} \frac{1}{\omega C_1} \right) \left( \alpha L_4 - \frac{1}{\omega C_1} \right) + \frac{3 \xi_{D31}^2 \omega^2 I_0^2}{4a^2} \right]^{1/2} \times (R I_0 - \frac{3 \xi_{D31}^2 \omega^2 I_0^2}{4a^2})^2
\]  

(25)

Comparing this equation with Eq. (23), the loss term including \(\xi_{D31}^{\prime\prime}\) is substituted for the term composed of \(R_1\) and \(I_0\). Since \(\xi_{D31}^{\prime\prime}\) is a negative value, \(-\frac{3 \xi_{D31}^2 \omega^2 I_0^2}{4a^2}\) in this loss term is positive and increases in proportion to \(I_0^3\). This means that the effective resonance resistance, \(R_1^3\), increases with the increase of \(I_0\). The ratio of the voltage to current at the drop point corresponds to
\( R' \). Defining an angle frequency of the drop point as \( \omega_D \), \( R' \) is given by
\[
R' = R_l - \frac{3\xi D_{31}' \theta}{4\omega_D l^2} f_0^2.
\] (26)

Solving for \( \xi D_{31}' \), we get
\[
\xi D_{31}' = \frac{32 f_0^2 (R_l - R')}{3l^2}.
\] (27)

Here, \( f_0 \) is the driving frequency at the drop point. Figure 9 represents the \( \xi D_{31}' \) values calculated with Eq. (27). Although the values slightly increased with the increase of the applied field, the significant changes were not observed. This suggests that we can treat the \( \xi D_{31}' \) as a material constant, which is obtained by substituting \( \xi D_{31}' \) into Eq. (3). Figure 10 shows the current-jumping curves in the (41/59) sample obtained by substituting \( \xi D_{31}' \) value into Eq. (25). The calculated curve reproduces exactly the experimental values including the jumping point and the drop point of the current-jumping phenomena.

The phase delay of the nonlinear domain-wall movements is actually confirmed by observing the waveform of the nonlinear voltage generated in the sample. Figure 11 represents the phase relation between the sample current and the nonlinear voltage in the (41/59) sample driven by a constant current. The bold solid curve and the open circles show the driving current and sample voltage waveforms, and the thin solid curve is the waveform of the third harmonic voltage obtained by the waveform separation from the waveform of the sample voltage. The broken curve indicates the waveform of another third harmonic voltage calculated by assuming that the phase delay does not exist, namely, that \( \xi D_{31}' \) is equal to zero. The thin solid waveform of the third harmonic voltage, which reflects the nonlinear movements of the domain walls, showed a slight phase delay, \( \theta' \), comparing with the calculated waveform. The \( \theta' \) indicates the phase delay of \( \xi D_{31} D_{33}' \) from \( D_{33} \) in Eq. (1), namely, indicating the phase delay of \( \xi D_{31}' \) form \( \int f_0 \) in Eq. (2). The \( \theta' \) is accordingly expressed by Eq. (28).
\[
\theta' = \sin^{-1}\frac{\xi D_{31}'}{\xi D_{31}}\left(D_{33} - \xi D_{31}' \int f_0 \right) \quad \text{(28)}
\]

Substituting \( \theta' \) into Eq. (28), \( \xi D_{31}' \) values are obtained. The broken line in Fig. 9 shows the absolute value of the \( \xi D_{31}' \) (1.1 × 10^21 V/C^3) calculated in this manner. The measurement error is probably large because of the small \( \theta' \) (-4°), however, this value is close to the value (0.97 × 10^21 V/C^3) calculated from the drop point of the current-frequency spectrum in the (41/59) sample driven at 4 V/mm shown in Fig. 9. This fact experimentally demonstrates that the large loss observed in the samples driven by a large current is caused by the phase delay of nonlinear movements of the non-180° domain walls.

5. Conclusions

(1) The curve calculated with our old simulation using only the nonlinear piezoelectric coefficient of third-higher term for electrical field, \( \xi D_{31} \), did not correspond to the experimental values of the current-jumping and the change of the resonance frequency measured in the PT-PSN samples having the crystal structure of rhombohedral phase.

(2) Introducing the nonlinear coefficient of third-higher term for mechanical stress, \( \xi D_{31} \), the discrepancy between the calculated values and the measured ones of the current-jumping can be corrected. Furthermore, adding the contribution of \( \xi D_{31} \) to our
old calculation, the calculated values of the change of resonance frequency exactly corresponded to the experimental values.

(3) Larger $\zeta_{31}$ was observed in the region of rhombohedral phase.

(4) Considering the nonlinear loss term by defining the complex $\zeta_{31}$, the measured curves of the current-frequency spectra were exactly reproduced by the calculated curves. This nonlinear loss term is due to the phase delay of the nonlinear movements of the non-180° domain walls.

References


