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Observation of piezoelectric resonance in time domain transient current of ferroelectric ceramics and crystals

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An abnormal resonance behavior was observed from the transient current in the time domain for both ferroelectric ceramics and single crystals excited with a stepwise electric field. The phenomenon can be accounted for as piezoelectric resonance. Corresponding resonant peaks in the frequency spectrum of impedance verified the piezoelectric origin of the abnormal resonance. Using a *RLC* series equivalent circuit of the piezoelectric resonance model, the resonance behavior of the transient current can be calculated and well fitted with experiments. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2031943]

Investigation of the transient current in ferroelectrics has an important significance in understanding the dielectric relaxation mechanisms[1–3] and domain dynamics.[4–7] Relaxation time distribution and domain reversal processes can be reflected in the transient current. Because of the potential applications,[8–10] both relaxation mechanisms and domain dynamics of the ferroelectric thin films have been widely discussed in terms of transient currents in the last decade.[1–3,6,7] For bulk ferroelectrics, such as ceramics and single crystals, the emphasis of the research has been on the domain dynamics through examination of the transient current in the time domain.[4,5,11,12] However, little attention has been devoted to the transient current in the study of dielectric relaxation. In order to investigate the dielectric relaxation by transient current in bulk ferroelectrics, a novel test system was built, and the transient current was characterized. The test system was capable of applying a 300 V stepwise electric field with a rise time about 50 ns, which cannot be realized by the traditional method.[11,12] During characterization of a ceramic sample, an abnormal resonance in the transient current was observed. This abnormal resonance behavior was also found to be present in a number of ferroelectric ceramics and single crystals under similar test conditions. An explanation for this behavior based on the model of piezoelectric resonance was developed, and supported with the experimental results. In this letter, we will report this interesting phenomenon along with our explanation for its presence.

Polycrystalline specimens of $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.32PbTiO_3$ (PMNT68/32, $E_c \sim 4.6$ kV/cm, diameter of 10 mm and thickness of 0.5 mm), modified $(1-x)PbZrO_3$-$xPbTiO_3$ (PZT, $E_c \sim 9$ kV/cm, square plate with width of 4 mm and thickness of 0.5 mm) and $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN, paraelectrics at room temperature, diameter of 10 mm and thickness of 0.5 mm) were prepared by the conventional ceramics processing method. In addition to the ceramics, single crystals of $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.32PbTiO_3$ (PMNT68/32, $E_c \sim 2.4$ kV/cm, square plate with width of 4 mm and thickness 0.5 mm) were grown using the Bridgman method in this study. The orientation of the PMNT single crystal sample was along the pseudocubic ⟨001⟩ direction which was determined by XRD (Rigaku D/max-2400). For the transient current measurements, a special test circuit was designed (Fig. 1), in which an *N*-channel metallic oxide semiconductor field effect transistor (N-MOSFET) was employed as a high-speed switch. When the N-MOSFET switch was driven by the MOSFET driver chip IR2110 (International Rectifier, IR), this circuit could supply a 300 V stepwise electric field with a rise time of about 50 ns. The capacitor in this circuit was used to enhance the power of the voltage source. When the N-MOSFET was triggered on, the instantaneous power could reach 1200 W. R3 in Fig. 1 is a bypass resistor, used as a discharge loop of the test sample. When the switch was shut down, a 50 Ω resistor was connected in series with the specimen in order to obtain the value of the transient current. The voltage across the resistor was measured using a Tektronix (TDS724C) oscilloscope. Prior to measurement, the specimens were polarized, and the electric field during measurement was applied parallel to the polarized field direction. The electric fields ranged from 2 to 6 kV/cm.

According to Merz's experiment,[4] there should be an exponential decay in the transient current when the stepwise field is applied parallel to the polarizing direction. This current is commonly referred to as the "displacement current" and obeys the exponential expression:

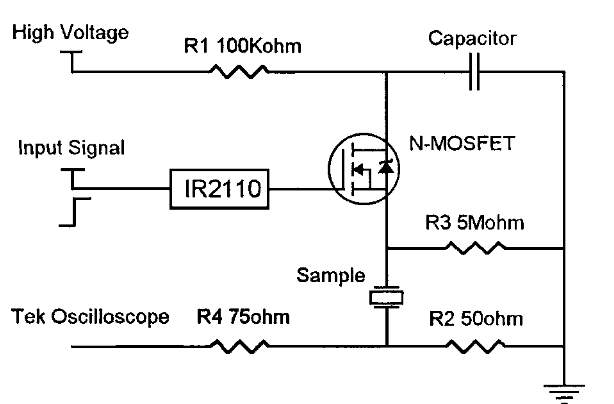

FIG. 1. Schematic circuit diagram of test platform. The current is measured across a series resistor, using an oscilloscope operated in the time capture mode.

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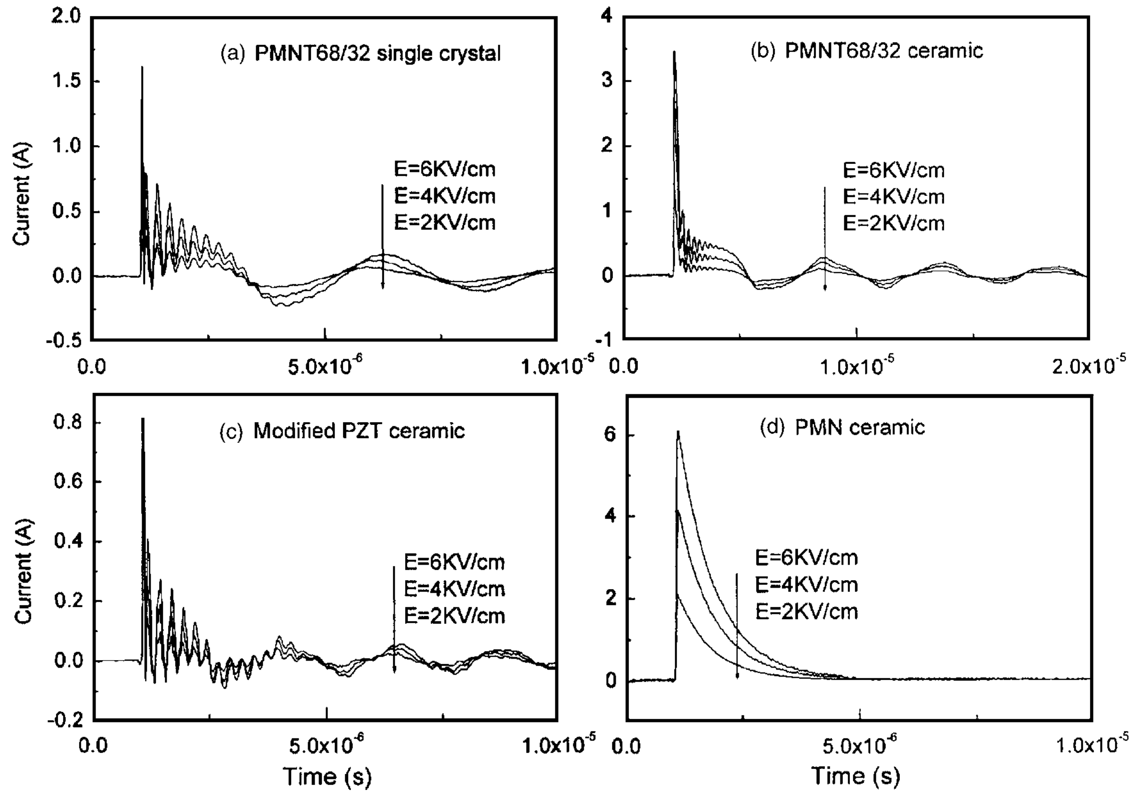

FIG. 2. Transient currents as a function of time for various ferroelectrics. (a) PMNT 68/32 single crystal, (b) PMNT 68/32 ceramic, (c) modified PZT ceramic, (d) PMN ceramic.

$$I(t) = A \exp(-t/\tau), \tag{1}$$

where $I(t)$ is transient current, A is amplitude, and τ is characteristic time. However Figs. 2(a)–2(c) all show abnormal transient currents when a stepwise electric field is applied to the specimens. It is clear that there are two different resonances, which are evident in the transient current shown in Figs. 2(a)–2(c). One is a high frequency resonance at the beginning of the current, while the other is a low frequency resonance. The resonance behaviors are similar for different samples, each exhibiting the same features. The abnormal resonances in transient current were found in both the ceramics and single crystals. The abnormal features are discussed in more detail for the modified PZT ceramic. Figure 2(c) shows the transient current of PZT ceramic when a stepwise field was applied in various amplitudes. Similar resonant frequencies can be observed with differences in resonant amplitude. Figure 2(d) is an exception of PMN ceramic, where the transient current decayed exponentially, which is normal for displacement current. No other resonance features can be observed in this case.

To explain what was observed, attention is focused on the piezoelectric resonance induced by the stepwise electric field. According to the theory of Fourier analysis,[13] a series of sine waves can be substituted for a square wave. The upper limit frequency of these sine waves is equal to the reciprocal of the rise time of the square wave, and the lower limit frequency is equal to the reciprocal of the width of the square wave. Therefore a stepwise wave can be substituted for a square wave, where the width of this square wave is infinity. In practice, when applying a stepwise field with a short rise time, it is equivalent to applying a series of sine waves to the specimen. The frequency of those sine waves is from 0 to 20 MHz, and the piezoelectric resonance was induced using the stepwise electric field. The stepwise electric field initially induced a mechanical vibration due to the converse piezoelectric effect, which generated a feedback to the current by the direct piezoelectric effect.

When the applied ac electric field has the same frequency as the resonant frequency of the specimen, traditional piezoelectric resonance is induced. Cady[14] calculated the current, which was induced by an ac electric field, named it "piezo current," and pointed out that the piezo current vibrates periodically. However in this experiment, a stepwise field induced two different piezoelectric resonances simultaneously, and the current decayed with these vibrations.

The resonance frequency of the modified PZT ceramic can be estimated from Fig. 2(c). The high frequency vibra-

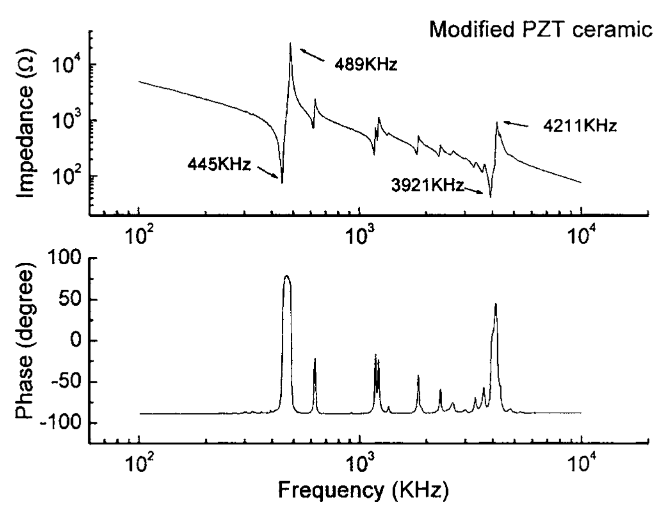

FIG. 3. Impedance of modified PZT ceramic as a function of frequency.

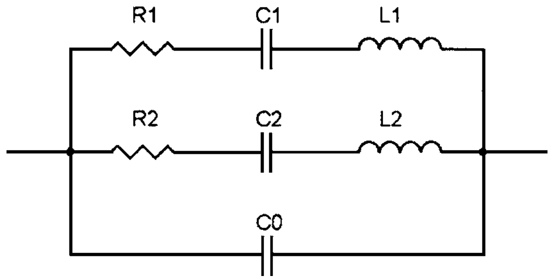

FIG. 4. Equivalent electrical circuit of a ferroelectric specimen.

tion period is about 0.24 μs and the low frequency vibration period is about 2.22 μs. Therefore the corresponding frequencies are on the order of 4.167 MHz and 450.45 kHz, respectively. Figure 3 shows the impedance as a function of frequency, which was measured using an HP 4192A impedance analyzer. It is clear that there are two resonant frequencies, 445 and 3921 kHz. For modified PZT ceramic, the frequency constant is about 2 kHz m. Thus the resonance frequency of 445 kHz corresponds to the planer vibration mode, and the resonance frequency of 3921 kHz is the thickness vibration mode.

In a series *RC* circuit, the charging current is calculated using Eq. (1). The electrical equivalent of a piezoelectric resonator around its resonant frequencies can be simulated as a *RLC* series circuit. In a less-damped condition, i.e., $R < 2(L/C)^{1/2}$, the piezoelectric resonance current in the *RLC* branch can be written as[15]

$$I(t) = A \exp(-t/\tau)\sin(\omega t + \mathrm{phi}), \tag{2}$$

where A is amplitude, which is dependent on the voltage applied and the parameter of the specimen. τ corresponds to the characteristic time, ω is the angular frequency, and equal to $2\pi f$, and Phi is a phase shift. Figure 4 shows the electrical equivalent circuit of the modified PZT ceramic. The transient current should contain three parts, one displacement current and two piezoelectric resonant currents. The total current can be written as

$$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)\sin(2\pi f_1 t + \mathrm{Phi}_1) + A_3 \exp(-t/\tau_3)\sin(2\pi f_2 t + \mathrm{Phi}_2), \tag{3}$$

where f_1 and f_2 are the resonant frequencies, 445 and

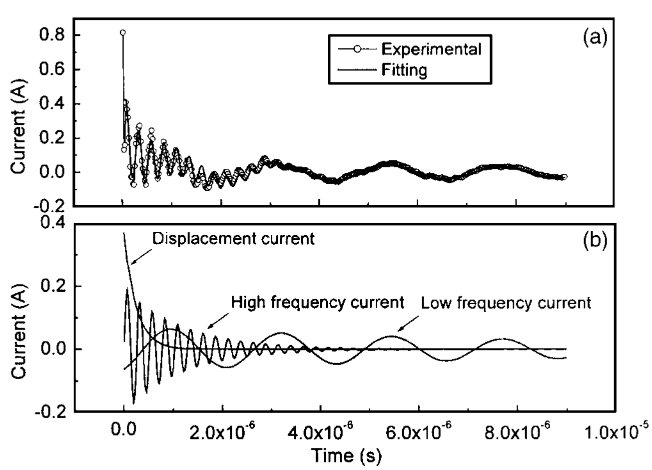

FIG. 5. Experimental and fitting data of modified PZT ceramic. ($E=6$ kV/cm) (a) total current of the fitting and experimental data, (b) three branches of total current.

TABLE I. Parameters from the fitting of Eq. (3) to the modified PZT ceramic. (E=6 kV/cm).

Parameter	Fitting data
A_1	0.399 52
A_2	0.0707
A_3	0.19997
τ_1	$2.433\,74 \times 10^{-7}$
τ_2	$1.010\,22 \times 10^{-5}$
τ_3	1.1741×10^{-6}
Phi_1	5.152 35
Phi_2	−0.361 01

3921 kHz, respectively. Phi_1 and Phi_2 are the phase shifts because the piezoelectric resonances did not occur simultaneously. Figure 5 shows the fitting and experimental data of the modified PZT ceramic with a pulse field of 6 kV/cm. It is evident that the fitting data correlated to the experimental data very well. Table I shows the fitting parameters.

Based on the above-presented discussion, it is understandable that there was no vibration observed in transient current in Fig. 2(d), because the vibrations of the transient current were induced by piezoelectric effect, a ferroelectric ceramics in its paraelectric phase would not generate piezoelectric resonance. The piezoelectric coefficient d_{33} is taken as an index of the piezoelectricity, and the value of d_{33} for PMN at room temperature is zero, which illustrates our explanation.

In summary, abnormal vibrations in the time domain transient currents of various ferroelectric ceramics and single crystals were observed. The corresponding piezoelectric resonant frequencies in the frequency domain spectrum were identified, and a piezoelectric resonance model was developed and fit the experimental data well.

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