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Study of abnormal transient current of PMN-32%PT ferroelectric single crystal in time domain and frequency domain

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Abstract A special test circuit was designed to measure the transient current in time domain. The measurement was performed on $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMN-32%PT) single crystal. Abnormal transient current was observed in time domain. Two oscillations were observed in transient current. By means of discrete Fourier transform (DFT), two peaks were found in frequency domain, which were corresponded to the resonance frequencies measured by frequency domain method. This phenomenon can be attributed to piezoelectric resonance. The transient current was calculated and fitted well based on a piezoelectric resonance equivalent electrical circuit model. Experimental results confirmed that no oscillation can be observed in PMN-32%PT single crystal samples after thermal annealing, which also verified that the piezoelectric resonance was the origin of the abnormal transient current.

Keywords Transient current · Piezoelectric resonance · Time domain · Frequency domain · PMN-32%PT single crystal

1 Introduction

Transient current has been intensively investigated in study of domain reversal process and relaxation mechanism in ferroelectrics [1–6]. Square pulse or step-wise electric field is most commonly used in those studies. Recently, our group observed abnormal oscillations in transient current both in ferroelectric ceramics and single crystals using a specially designed test circuit to generate a step-wise electric field with a quick rise time. It is believed that those oscillations were excited by piezoelectric resonance [7]. In many practical applications, piezoelectric resonators are mostly driven by an AC electric field. Based on the above experimental phenomenon, using step-wise driving field may provide some new features to some of the applications.

Relaxor ferroelectric single crystals, such as $(1-x)Pb(Zn_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PZN-PT) and $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PMN-PT), have received much attention because of their excellent piezoelectric and dielectric properties [8–10]. In present work, a 0.68PMN-0.32PT (PMN-32%PT) single crystal was used as a typical material to study the abnormal transient current induced by step-wise electric field in time domain. Discrete Fourier transform (DFT) was performed to analyze the abnormal transient current. Compared to the frequency spectrum of the same sample, the authors verified that the abnormal transient current is stem from piezoelectric resonance.

2 Experimental

Single crystal of the PMN-32%PT was grown by the Bridgman method using accelerated crucible rotation technique (ACRT) [11]. Then the single crystal was cut into square plate with <001> orientation. The dimensions of the plate were 5×5 mm with a thickness of 0.4 mm. The sample was polarized along thickness direction before measurement.

In order to study the abnormal transient current excited by step-wise electric field, a special driving circuit was built as described in [7]. An N-channel metallic oxide semiconductor field effect transistor (N-MOSFET) was employed as a high-speed switch. Using this circuit, a step-wise voltage as

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high as 300 V is able to apply onto the sample in a quite short time (less than 100 ns). The direction of the exciting electric field was parallel to the poling direction. A Tektronix oscilloscope (TDS724C) was employed to record the waveform of the transient current in different time intervals. The impedance frequency spectrum of the sample was measured by a precision impedance analyzer (Agilent 4294A).

3 Results and discussion

Figure 1 shows the transient current of the sample in different time intervals from 5 to 20 μs. The transient current profiles increased when the electric field increased from 2.5 kV/cm to 7.5 kV/cm. Compared to displacement current, which usually decreased exponentially without oscillation, two abnormal oscillations were observed in the transient current. In Fig. 1(a), a high frequency oscillation was found at the beginning of the current. The oscillation frequency was calculated roughly with the time interval between two continuous peaks of the oscillations, and the result was about 4 MHz. Similar calculation was also performed for Fig. 1(c), and the corresponding frequency was about 200 kHz.

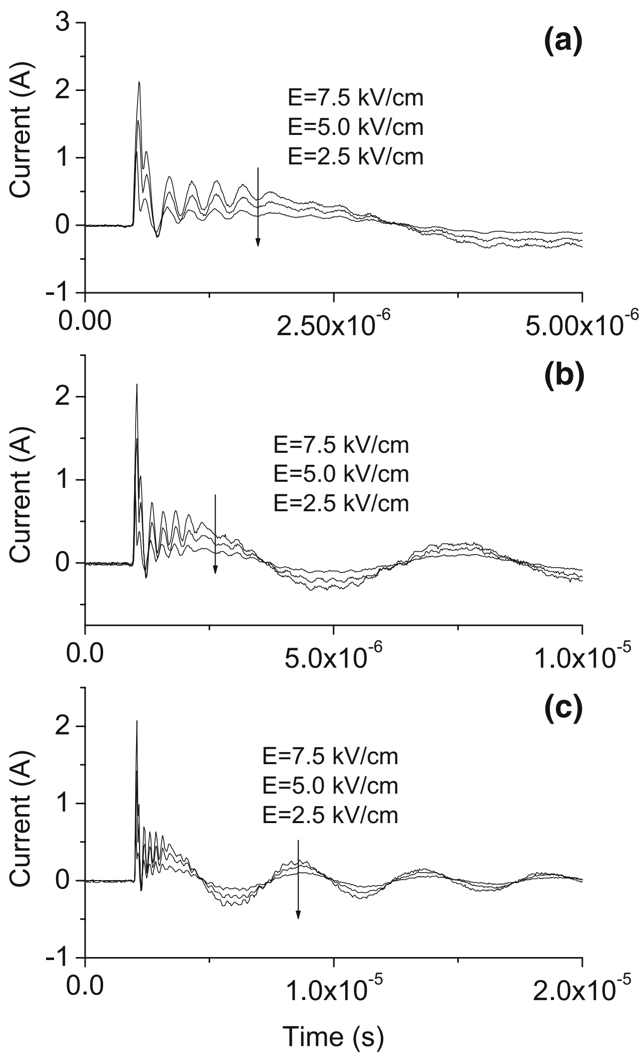

Fig. 1 Transient current of PMN-32%PT single crystal in different time intervals. (**a**) 5 μs; (**b**) 10 μs; (**c**) 20 μs

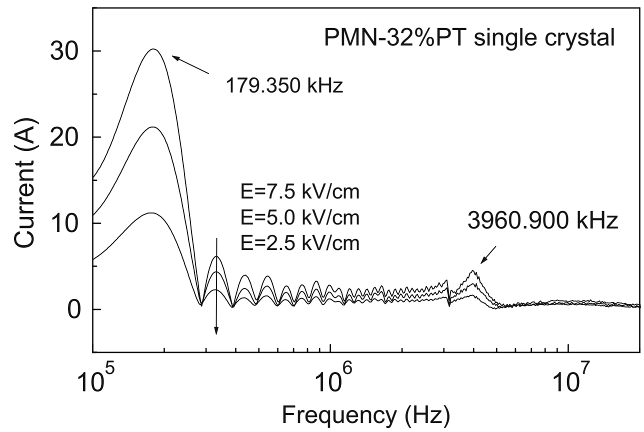

Fig. 2 Complex transient current of PMN-32%PT single crystal calculated by DFT with respect to frequency

Signals obtained in time domain can be analyzed in frequency domain through Fourier transform (FT) [12]. Because the data of transient current were discrete in time domain, a program was developed based on discrete Fourier transform (DFT). Though fast Fourier transform (FFT) could increase efficiency of computation, it would lose some subtle information in frequency domain especially at low frequencies. The following Eqs. 1, 2, and 3 were used to calculate complex transient current in frequency domain:

$$I'(\omega) = \sum_{t=0}^{t_{\max}} I(t)\cos(\omega t) \tag{1}$$

$$I''(\omega) = \sum_{t=0}^{t_{\max}} I(t)\sin(\omega t) \tag{2}$$

$$R(\omega) = \sqrt{I'(\omega)^2 + I''(\omega^2)} \tag{3}$$

where $I(t)$ is the transient current obtained in time domain, t is the time which begins from the maximum of transient current, $t_{\max}$ is the time length from the maximum to the end of transient current, and ω is angular frequency ($2\pi f$). $I'(\omega)$, $I''(\omega)$ and $R(\omega)$ represent real part, imaginary part and amplitudes of the complex transient current in frequency domain respectively. The data shown in Fig. 1(b) was selected to calculate the frequency spectrum, because in Fig. 1(b) both high frequency and low frequency oscillations were recorded clearly. The amplitudes of complex transient current with respect to frequency were shown in Fig. 2. The unit of abscissa is Hz, while A for ordinate. It

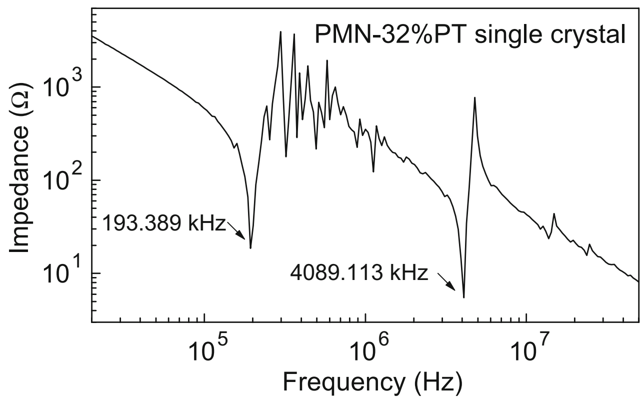

Fig. 3 Electric impedance of PMN-32%PT single crystal with respect to frequency

can be observed that the current amplitudes increased with electric field strength. Two peaks were found at 179.350 and 3,960.900 kHz. These two frequencies shown in Fig. 2 are close to the frequencies calculated from Fig. 1. When the voltage is fixed, the current reaches its maximum while the impedance reaches its minimum. Therefore a hypothesis is introduced that the electric impedance should reach its minimum at frequencies of 179.350 and 3,960.900 kHz. Figure 3 shows dependence of the electric impedance of PMN-32%PT single crystal on frequency. There are two peaks with frequencies of 193.389 and 4,089.113 kHz. A plate ferroelectric sample contains two piezoelectric resonance modes, i.e. the planar mode and the thickness mode [13]. Thus 193.389 kHz is planner mode resonance frequency while 4,089.113 kHz is thickness mode resonance frequency. There are some irregular vibrations between those two frequencies in Fig. 3, which may be caused by geometric discrepancy between the width and length.

Compared with the frequencies obtained in Figs. 2 and 3, the abnormal transient current seems to be induced by piezoelectric resonance. Based on Fourier analysis, a square wave can be substituted by a series of sine waves. Piezoelectric resonance could be excited by those sine waves through converse piezoelectric effect, when those sine wave frequencies are equal to the resonance frequency. Once the resonance was excited, there is an oscillation feedback to the transient current through piezoelectric effect. In the present work, the rise time was less than 100 ns, thus the upper limit frequency was more than 10 MHz. The two resonance frequencies of the sample were below this frequency. This ensured that those two resonances could be excited.

Table 1 Fitting parameters of the PMN-32%PT single crystal data to Eq. 5 (E=7.5KV/cm).

Parameter	Fitting data
A_1	0.84728
A_2	0.4438
A_3	0.84874
τ_1	3.0991×10^{-7}
τ_2	1.0569×10^{-5}
τ_3	4.0668×10^{-7}
phi_1	−0.044
phi_2	12.49162

The equivalent electrical circuit of a piezoelectric resonator is a *RLC* series circuit. When a step-wise electric field is applied to the sample, the current could be calculated using Eq. 4 as mentioned in [7]:

$$I(t) = A\exp(-t/\tau) \times \sin(2\pi ft + \text{phi}) \tag{4}$$

where A is amplitude of transient current, which is dependent on the applied voltage and the parameter of the

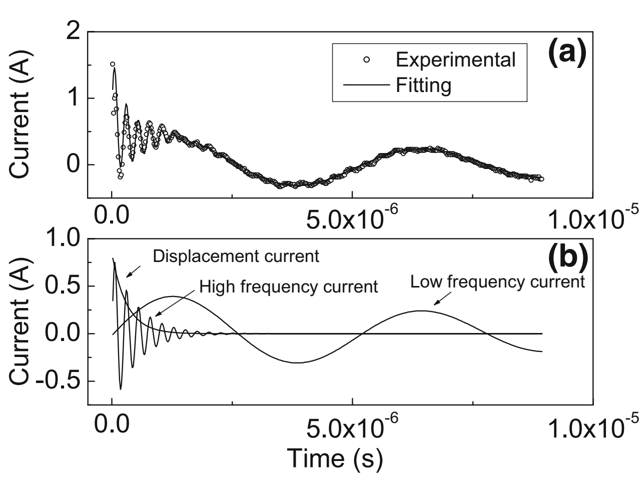

Fig. 4 Experimental and fitting data of PMN-32%PT single crystal (E=7.5 kV/cm). (**a**) Experimental current and total fitting current; (**b**) three branches of total fitting current

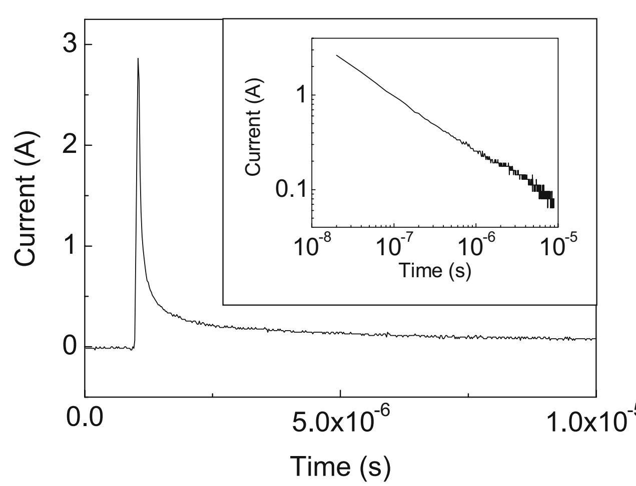

Fig. 5 Transient current of the PMN-32%PT single crystal after annealing (E=7.5 kV/cm). *Inset* Transient current displayed in logarithmic coordinate from maximum current

specimen, τ is corresponding to the characteristic time, f is oscillation frequency and phi is phase shift. In the present work, two oscillations were found in the transient current. The equivalent electrical circuit of the sample should contain three branches. Two RLC series branches represent two piezoelectric resonances and a capacitor represents a normal static capacitor of the sample. Equation 5 was used to fit the experimental data:

$$\begin{aligned} I(t) = & A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \\ & \times \sin(2\pi f_1 t + \mathrm{phi}_1) + A_3 \exp(-t/\tau_3) \\ & \times \sin(2\pi f_2 t + \mathrm{phi}_2) \end{aligned} \tag{5}$$

where f_1 and f_2 are the resonance frequencies. In this experiment they are 193.389 and 4,089.113 kHz, respectively. Figure 4 shows the fitting and experimental data of the sample with the electric field of 7.5 kV/cm. It is evident that the data fitted the experimental data well. Table 1 gives the fitting parameters.

There is no piezoelectricity of PMN-32%PT single crystal when the sample was annealed from 473 K with a rate of 3 K/min (T_c=416 K). Figure 5 shows the transient current of the sample after annealing. It can be seen that the transient current decreased without oscillation. However, the current did not decrease exponentially shown in the inside figure of Fig. 5. For a linear dielectric material without piezoelectricity, transient current usually obeys the exponential law. For a ferroelectric material, the sample behaves as linear dielectric and nonlinear ferroelectric simultaneously. Therefore, the current shown in Fig. 5. should not obey the exponential law. Under the excitation of the stepwise field, the doped sample would be poled by the stepwise field through the reorientation of domains. When a second stepwise field was applied successively, the transient current would be similar to the current shown in Fig. 1. Oscillation of the transient current reappeared. These results also indicated that the abnormal oscillations were origin from piezoelectric resonance.

4 Conclusion

Abnormal transient current was observed in PMN-32%PT single crystal when a step-wise electric field was applied with a short rise time (less than 100 ns). The transient current was analyzed by means of DFT. Two characteristic frequencies observed through DFT were close to the resonance frequencies measured by frequency domain method. It is suggested that the abnormal transient current was induced by piezoelectric resonance, and it was calculated with an equivalent electrical circuit of piezoelectric resonance model. Transient current without oscillations of PMN-32%PT single crystal after annealing also supported the conclusion.

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References

1. W.J. Merz, Phys. Rev. **95**, 690(1954)
2. V. Shur, E. Rumyantsev, S. Makarov, J. Appl. Phys. **84**, 445 (1998)
3. U. Belegundu, K. Uchino, J. Electroceram. **6**, 109 (2001)
4. T. Horikawa, T. Makita, T. Kuroiwa, N. Mikami, Jpn. J. Appl. Phys. **34**, 5478 (1995) Part 1
5. J.D. Baniecki, R.B. Laibowitz, T.M. Shaw, P.R. Duncombe, D.A. Neumayer, D.E. Kotecki, H. Shen, Q.Y. Ma, Appl. Phys. Lett. **72**, 498 (1998)
6. A.K. Jonscher, *Dielectric Relaxation in Solids* (Chelsea Dielectrics, London (1983)
7. L. Jin, X. Yao, X.Y. Wei, Z.Z. Xi, Appl. Phys. Lett. **87**, 072910 (2005)
8. S.E. Park, T.R. Shrout, J. Appl. Phys. **82**, 1804 (1997)
9. R.F. Service, Science **275**, 1878 (1997)
10. H. Fu, R.E. Cohen, Nature **403**, 281 (2000)
11. Z.R. Li, Z.Z. Xi, Z. Xu, X. Yao, J. Mater. Sci. Lett. **21**, 1325 (2002)
12. A.V. Oppenheim, A.S. Willsky, I.T. Young, *Singles and Systems* (Prentice Hall, Englewood Cliffs (1983)
13. ANSI/IEEE STD. 176–1987, IEEE Standard on Piezoelectricity (IEEE, New York, 1987)