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FAST TRACK COMMUNICATION

Evolution of piezoelectric resonance in switching current of $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ ferroelectric single crystal excited by a stepwise electric field

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Abstract

Piezoelectric resonance current and switching current of $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMT–32%PT) single crystal were investigated by a stepwise electric field. As the reversely applied field increased from the coercive field to higher levels, high frequency resonance current always existed while the low frequency resonance current was replaced by a peak in the switching current gradually. This phenomenon is discussed based on a domain reversal process. The maximum switching current observed in switching current is fitted well using the equation proposed by Merz. This indicates that domain wall growth, instead of domain nucleation, plays an important role in that process.

1. Introduction

A crystal is said to be a ferroelectric when it exhibits a spontaneous polarization, even in the absence of an electric field, and the direction of this polarization can be reversed by an external electric field [1]. Ferroelectric random access memories (FeRAMs) are fabricated based on such a polarization reversal process [2]. The polarization switching dynamics is important to the application. Usually this process is investigated by switching current, for example, in $BaTiO_3$ single crystals and other ferroelectrics [3–5]. When square pulses parallel or antiparallel to the polarization direction of samples are applied, displacement current and switching current can be observed through an oscilloscope. Maximum switching current i_{max} and switching time t_s can be determined from this experiment as shown in the inset of figure 1(a). This switching process will be dominated by domain nucleation or domain wall motion separately according to the strength of the applied field. Subsequently, this method was adopted by other groups [6–11], and domain reversal dynamics was developed by Ishibashi and Takagi [8], Shur *et al* [9] and Viehland and Li [10] based on the switching current.

On the other hand, a ferroelectric possesses piezoelectricity when it is poled. Polarization switching process is a depolarization and reorientation process, which would weaken or eliminate the present polarization state. Furthermore the piezoelectric properties are dependent on the polarization state. Thus it is interesting to investigate the variation of the piezoelectric properties when polarization is reversed. Polarization reversal is a very quick process that would complete in the order of a microsecond, whereas the piezoelectric properties are usually measured at pseudo-static condition. Recently, our group observed abnormal piezoelectric resonances in transient current of both ferroelectric ceramics and single crystals,

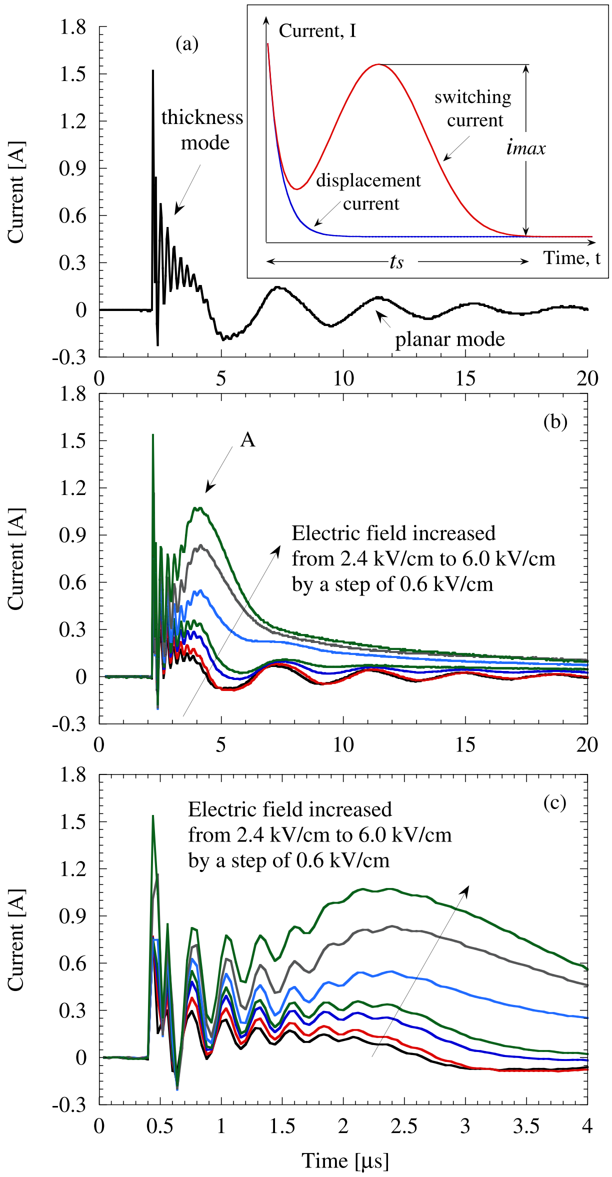

Figure 1. Transient current of PMN–32%PT single crystal. (*a*) Electric field parallel to polarization; only the current measured at 6 kV cm^{-1} is shown. Inset displays a schematic diagram of displacement current (field parallel to polarization) and switching current (field antiparallel to polarization); (*b*) electric field antiparallel to polarization and strength increased from 2.4 to 6 kV cm^{-1}; (*c*) local profile of transient current shown in figure 1(*b*). (This figure is in colour only in the electronic version)

when excited by a stepwise field [12]. Then this universal phenomenon in different electromechanical coupling modes was proved [13]. The authors also discovered that there is no piezoelectric resonance in non-ferroelectrics or ferroelectrics in their paraelectric phase excited by the stepwise field [12]. Therefore, the appearance of piezoelectric resonance can be considered as an index of the piezoelectricity at a certain level. By this method, domain reversal process and piezoelectric resonance could be studied simultaneously through transient current. This is important in understanding the relationship between the piezoelectricity and ferroelectricity, when the polarization state is reversed. In this communication, we investigated the evolution of piezoelectric resonance in the switching current of $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMN–32%PT) single crystals and discussed the results based on the model proposed by Merz [3].

2. Experimental

The test system has been described in [12]. By this system, a stepwise field can be applied to the sample with a rise time less than 50 ns and the maximum output voltage is 300 V. In order to study the domain reversal process through this system, the ferroelectric materials should possess a low coercive field and relatively large remanent polarization. The $xPb(Mg_{1/3}Nb_{2/3})O_3$–$(1-x)PbTiO_3$ (PMNT) system is highly suitable for this investigation [14]. The PMN–32%PT single crystal was grown using an accelerated crucible rotation technique (ACRT) and Bridgman method, which was oriented along pseudocubic ⟨0 0 1⟩ directions by XRD. The sample was cut into a square plate with a width of 4 mm and a thickness of 0.5 mm. Silver electrodes were plated on the main surface and fired at 850 °C for 30 min. Before applying the field, the sample was poled at a high electric field in hot silica oil (6 kV cm^{-1}, 80 °C for 30 min). A step field was first applied on the sample in the direction of polarization and then another step field was applied in the opposite direction. Similar processes were repeated for the same sample but with different strengths from 2.4 to 6 kV cm^{-1}. The coercive field is 2.4 kV cm^{-1}, which was determined from the hysteresis loop at 1 Hz.

3. Results and discussion

In the normal case, transient current consists of two piezoelectric resonance currents when the electric field direction is parallel to the polarization direction (figure 1(*a*)). Those two resonances are identified as planar mode (low frequency) and thickness mode (high frequency) piezoelectric resonances [12, 13]. When a field antiparallel to the polarization is applied to the sample, transient current displays another profile as depicted in figures 1(*b*) and (*c*). This profile can be divided into three steps according to the field strength. At 2.4 and 3.0 kV cm^{-1}, two vibrations similar to those shown in figure 1(*a*) were observed (figure 1(*c*)). These curves are typically piezoelectric resonance current. From 3.6 to 4.2 kV cm^{-1}, a visible peak indicated at the A site emerged and the low frequency resonance disappeared gradually. At a higher level (larger than 4.8 kV cm^{-1}), only the high frequency resonance, which is not affected by the field, is observed. The low frequency resonance was substituted by the peak at the A site completely. There is an evident transition from piezoelectric resonance to domain switching when the field was increased.

Why does the low frequency disappear when the field is increased, while the high frequency resonance still exists? To answer this question, the polarization state should be taken into account. Because the polarization state could be changed through the domain nucleation and the domain wall motion. When an antiparallel field is applied, the net polarization weakens then decreases to zero and finally reaches a negative value compared with its original value. The precondition is that the field strength is larger than the coercive field. In this process, domain nucleation occurs at first and is then followed by domain wall growth along sidewise motion [3–7]. When a negative polarization is acquired, the step field is already changed into a steadily static field instead of an ac field; thus the low frequency piezoelectric resonance would not be excited. To the planar mode, the piezoelectric resonance featured by a high frequency vibration attenuates in a relatively short time period (stopped at around 4 μs in figure 1(*b*)), whereas the net polarization is still kept at a positive value. Thus piezoelectric resonance can be excited by the step field even though the polarization state has been changed already. It is noted that all the steps of domain reversal are time related; therefore different responses to the antiparallel field could be reflected in the transient current.

When the applying field is equal to the coercive field, the polarization should be zero. In this case there should be no piezoelectricity in the material. However in figure 1(*b*) (from 2.4 to 4.2 kV cm^{-1}), low frequency resonance is still observed. The following conjecture could help us to understand this phenomenon. Usually the coercive field is determined through the hysteresis loop by applying an ac field (sinusoidal or triangular function), and the results indicate that the coercive field will increase by increasing the frequency of the ac field [3, 15, 16]. With increasing frequency the nucleation and growth of domain wall cannot follow the change in the field; therefore higher voltages are required for polarization reversal [15]. As shown in figure 2, polarization reversal will occur from point 4 to 1 or 2 to 3 in a period. Increasing frequency is equal to shortening the time between points 4 and 1. The stepwise electric field represented by a dashed line in figure 2(*a*) can be divided into two parts. The first part of this field from point 4 to 1 is important to reversal, which is similar to a triangular field in its first one-fourth periods. Thus a stepwise field with a short rise time (50 ns) can be considered a high frequency ac field between points 4 and 1. In order to get a net zero polarization by this field, a higher voltage is expected. In figure 1(*b*), the low frequency resonance was weakened when the field is $\geqslant$3.6 kV and disappeared completely at 4.8 kV cm^{-1}. Compared with the current at low field (at 3.0 and 3.6 kV cm^{-1}), it is clear that the amplitude of the low frequency resonance at 3.6 kV cm^{-1} is much lower than that at 3.0 kV cm^{-1} (from 5 to 20 μs). Only one or two periods are observed. This indicates a transition, but it is not clear enough to determine the coercive field through the switching current.

In the following we will determine the coercive field through the relationship between maximum switching current i_{max} and the stepwise field E suggested by Merz [3]. Compared with the data obtained by Merz, the peak at the A site corresponds to i_{max} (inset of figure 1(*a*)). There are two equations to describe the relationship between i_{max} and E. At low field, domain nucleation dominates the current profile, $i_{max} = a\mathrm{e}^{-\alpha/E}$ with a = constant. At high field, domain wall growth dominates the profile. In this situation,

$$i_{max} = a(E - E'), \qquad (1)$$

where E' is a kind of coercive field strength. When the field is in excess by 3.6 kV cm^{-1}, i_{max} can be acquired in figure 1(*b*).

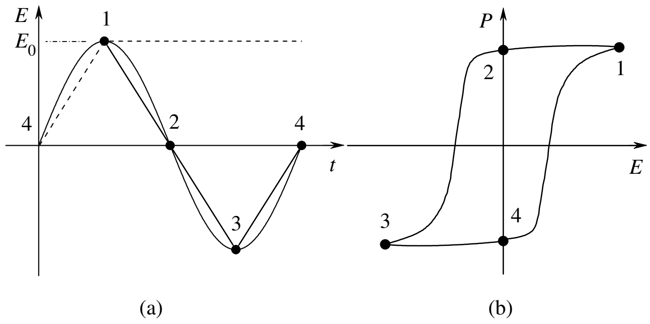

Figure 2. Relationship between the ac field and the hysteresis loop. (*a*) Ac sinusoidal or triangular field in a period, and broken line represents a stepwise electric field; (*b*) typical hysteresis loop of a ferroelectric. The polarization states denoted by points 1, 2, 3, 4 in figure 2(*b*) correspond to the electric field in figure 2(*a*).

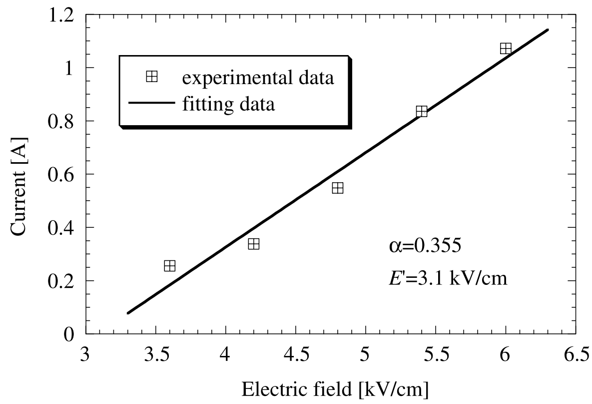

Figure 3. Maximum switching current i_{max} versus applied field E. The points represent experimental data and the line corresponds to the fitting using equation (1).

Therefore only five i_{max} between 3.6 and 6.0 kV cm^{-1} were used to fit. Based on the above discussion, domain nucleation does not influence the low frequency resonance. When the field changed between 3.6 and 6.0 kV cm^{-1}, the low frequency resonance disappeared gradually. This indicates that domain wall motion plays an important role in this process. Usually domain reversal is accomplished by sidewise motion of 180° domain walls [6, 7]. However using the switching current method, we could not identify whether there are some contributions by the motion of 90° domain walls. After all, domain reversal process in this communication is studied by a macroscopic method instead of a microscopic one. Figure 3 displays a well fitting result by equation (1) (correlation coefficient = 0.985) and $E' = 3.1$ kV cm^{-1}. This value, which is larger than the coercive field (2.4 kV cm^{-1}) measured from the hysteresis loop, agrees with the above discussion. It is noted that due to the piezoelectric resonance in switching current, it is difficult to measure the t_s through the transient current. This will be discussed in a future work.

4. Conclusions

In summary, the piezoelectric resonance in switching current has been investigated by means of a stepwise electric field.

The high frequency resonance is independent of the reversal field, while the low frequency resonance is replaced by a peak in switching current with increasing field. This means that piezoelectricity does not disappear immediately but gradually in this process. The maximum switching current can be fitted well based on domain wall growth using the equation proposed by Merz.

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