

FAST POLARIZATION CHANGES IN PZT CERAMICS

BY HIGH-VOLTAGE PULSES

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Abstract

Experiments on fast spontaneous polarization changes in PZT ceramics are described. The interactions between spontaneous polarization P_s and space-charge polarization $P_{sp.ch.}$ are discussed. The fast polarization changes are made with the aim of producing electron beam emission and acceleration from ferroelectric surfaces.

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INTRODUCTION

Some features of prepoled ferroelectrics, i.e. surface-charge densities of the order of 10^{14} cm^{-2} and strong electric fields due to spontaneous (P_s) and space-charge ($P_{sp.ch.}$) polarization¹, make these materials interesting for the field of particle accelerators and in particular as an electron-emitting source². Normally, both polarization components are mutually compensated (screened), but, when one of them is changed rapidly, short high-voltage spikes and electron emission can be expected during a brief time-interval. Polarization changes in ferroelectric materials can be induced by temperature changes, by external electric field pulses, or by mechanical pressure. The electric field method is discussed below in some detail. Significant transient effects are expected during full P_s reversal and phase transitions triggered through high-voltage pulses.

First experimental results are shown for $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ solid solutions (PZT) with a content of Ti up to 5% mol. The characteristic features of these materials are the phase sequence antiferroelectric-ferroelectric-paraelectric (AFE-FE-PE) when the temperature is raised, and a dependence of the phase width of the intermediate ferroelectric phase on the Ti content³, on the external electric field⁴, on the mechanical pressure⁵, and on vacancies in the oxygen and the lead sublattices forming F- and V-colour centres⁶. The F-colour centres dominate in oxygen-reduced PZT materials. They behave as acceptors, trapping one or two electrons (F_1 - and F_2 -colour centres, respectively). These carriers can be released by thermal or optical excitation ($F_2 \xrightarrow{1 \text{ eV}} F_1 + e^-$). They participate in P_s

FAST POLARIZATION CHANGES IN PZT CERAMICS

screening and bulk current generation, and may contribute to electron emission. In the case of metal-electroded samples, electrons injected from the cathode play an important role in $P_{sp,cb}$ formation and in P_s screening.

EXPERIMENTAL RESULTS

Silver electrodes were deposited on polished disk-shaped $Pb(Zr_xTi_{1-x})O_3$ ceramic samples, of thickness 0.5 to 2.0 mm. Thin wires provided the electric contact. The sample was placed inside a vessel containing silicone oil in order to facilitate temperature stabilization or regulation. The samples were connected in series with a condenser of 0.1 to 1 μF (Figure 1).

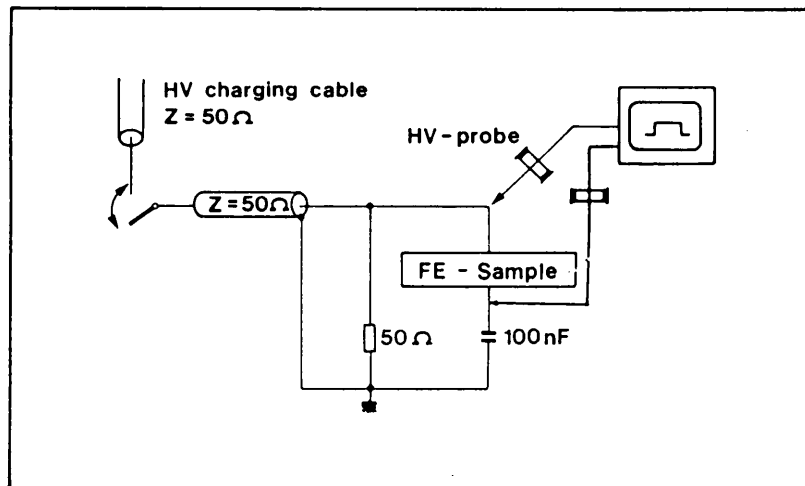


FIGURE 1 Simplified measuring circuit.

The PZT ceramic samples were prepoled under a field E_{dc} of 1 to 3 kV/cm applied in the PE state and cooled under the field down to the FE state. High-voltage rectangular pulses of alternating polarity, with a few nanoseconds rise-time, were applied to the samples at different temperatures, and the voltage across the sample was measured within or outside the intermediate ferroelectric phase.

Figure 2 shows the positive voltage pulses (with the same polarity as that of the bound surface charges P_s) measured on a $Pb(Zr_{0.99}Ti_{0.01})O_3$ sample at five different temperatures during cooling. The wave form at 225 °C (inside the FE state) is typical of fast P_s reversal. At temperatures of 240 °C and 195 °C, which are still outside but in the vicinity of the intermediate FE phase for zero electric field, a similar behaviour has been observed. After the voltage drops owing to P_s change, it recovers with a rise-time that is

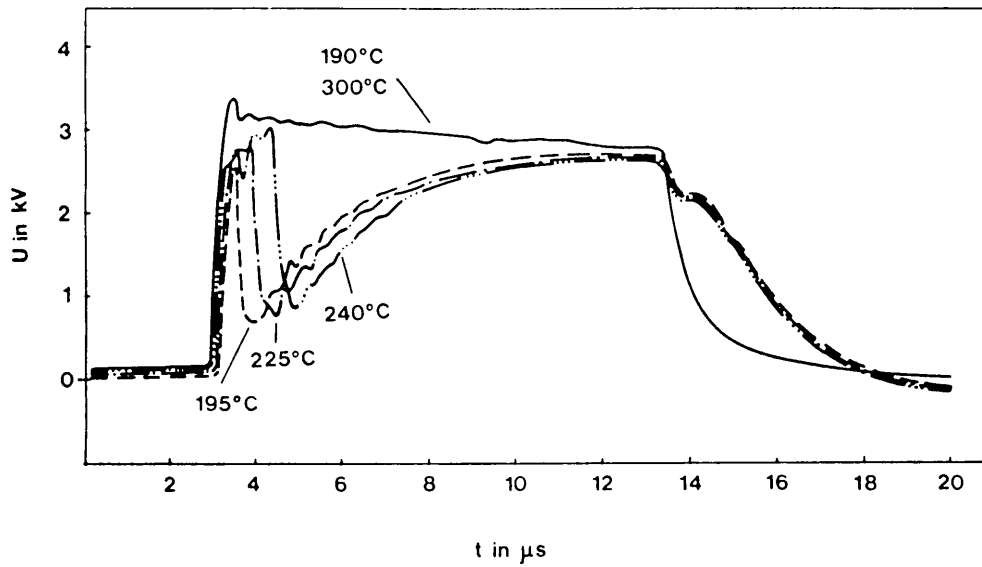


FIGURE 2 Voltage wave forms measured across a PZT sample of 1 mm thickness at different temperatures inside and outside the FE state.

determined by the filling time of the capacitor in series with the sample. At 300°C (far above the FE-PE phase transition temperature) and at 190°C (below the AFE-FE phase transition temperature) P_s changes are absent. The rectangular wave form is determined mainly by the 50 Ω matching resistor (Figure 1).

In Figure 3 a number of high-voltage wave forms (amplitude 50 kV/cm) are shown, measured on a PZT sample with 0.5% Ti during cooling from 256°C (PE state) down to the phase-transition temperature, where breakdown due to phase transition occurred.

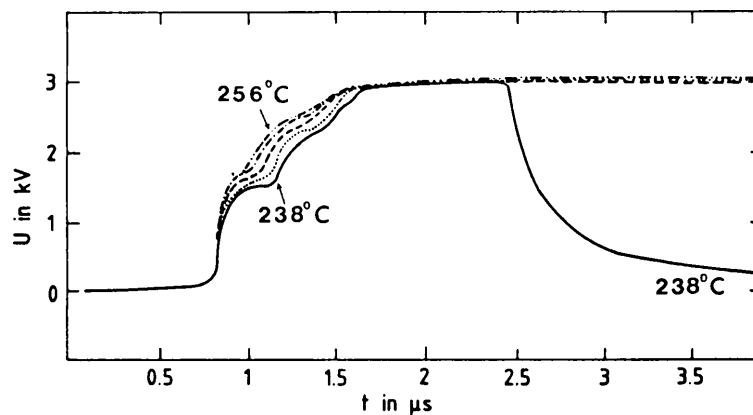


FIGURE 3 Variation in the rise of the HV pulse wave forms across a PZT sample during cooling from the PE state to the FE phase boundary.

FAST POLARIZATION CHANGES IN PZT CERAMICS

When the temperature was raised again to the PE state, the breakdown disappeared. In the PE phase, the pulse rise-time increases gradually with decreasing temperature.

The interaction of P_s and of $P_{sp.ch.}$ has been studied for prepoled PZT (4% Ti) samples under different conditions by measuring the response to the rectangular high-voltage pulses shown in Figure 4. Two series of HV pulses were applied successively within the FE phase; the first (1-3) with the same sign as that of the bound P_s charges and the next (4-6) with the opposite sign. The application of higher electric field pulses (> 50 kV/cm) of positive sign leads to P_s reversal (Figure 4b, wave form 9), which is often accompanied by surface flashover, by electrode damage, by breakdown through the bulk of the material, or by complete destruction of the sample.

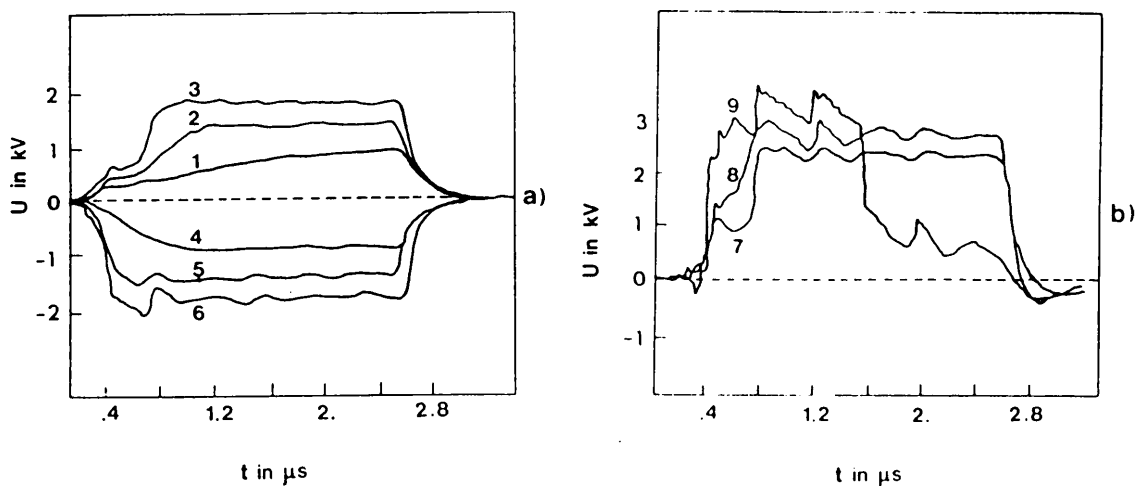


FIGURE 4 Voltage wave forms measured across a prepoled PZT sample within the FE phase as a response to several series of positive and negative short rectangular pulses. a) The amplitude is gradually increased during the first positive rectangular input pulses (1-3) as well as through the following pulses (4-6) with negative polarity. b) At high pulse amplitudes (7-9) the polarization P_s is finally reversed (9) with respect to the direction of the prepolarization current.

DISCUSSION

It has been demonstrated^{1,7} that the switching time of the P_s vector depends strongly on the sample thickness, on the temperature, on the applied electric field strength, the electrical conductivity, and the nature of the electrodes, on the state of the crystal surface and the presence of defects, and on the domain geometry. When a high electric field is applied to the prepoled sample in the direction opposite to its polarization, each existing antiparallel domain starts to grow with a very fast sideways and forward motion of domain walls¹. The proper selection of the above-mentioned conditions permits a rapid

H. GUNDEL, J. HANDEREK, H. RIEGE, E.J.N. WILSON AND K. ZIOUTAS

macroscopic P_s change, whereas the different components of $P_{sp.ch.}$ can only be changed slowly. Destruction of the sample and of the electrode may be caused by the very strong repulsion between suddenly reversed \vec{P}_s and by the liberated electron charges. The origin of these effects can be similar to that of the known fatigue effect⁸. It is not quite clear whether ionic or electronic processes play the dominant role. Probably the domain 'fusion'¹ has to be taken into account.

The field-strength amplitude that is necessary for the P_s reversal of a prepoled sample to take place within much less than 100 ns is one order of magnitude higher than the coercive field obtained from hysteresis loop measurements at 50 Hz with an a.c. field. The presence of defects and $P_{sp.ch.}$ tends to increase the coercive field¹. Rather high electric fields are also required for fast phase transitions (AFE \rightarrow FE or PE \rightarrow FE) in order to cross the phase boundary, even when the starting temperature differs only slightly from the phase-transition temperature at $E = 0$ (Figure 2).

SUMMARY

The achievement of spontaneous polarization reversal and of phase transitions by high-voltage pulses in far less than 100 ns opens up the way to interesting applications, especially in accelerator technology. The faster the polarization change the higher the local surface charge density and the electric field strength which can be expected during a short time-interval. One of the most important applications is electron emission and acceleration from FE surfaces. It is, however, necessary to find methods of preventing the destruction of the samples and of the electrodes. The next step is to master the electron emission process induced by spontaneous polarization reversal or by fast phase transition. The right ferroelectric materials, the right electrode type, and the most convenient geometrical, environmental, and circuit parameters have to be found. The large fields perpendicular to the surface layers can then hopefully be used to give the electrons an initial acceleration which will produce a beam of small emittance and high intensity. Laser illumination of the cathode surface layers of prepoled samples will cause additional electron liberation from F_2 -colour centres and can amplify the electron emission. A high-density electron cloud will be created inside the ferroelectric sample very near to the cathode surface, and at the same time a P_s reversal will be induced. The acceleration of the liberated electrons may then start under the combined action of the space-charge field and an external extraction field.

FAST POLARIZATION CHANGES IN PZT CERAMICS

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