

**FAST POLARIZATION CHANGES IN FERROELECTRICS
AND THEIR APPLICATIONS IN ACCELERATORS**

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Abstract

Several mechanisms are described which lead to fast polarization changes in ferroelectric materials. The resulting surface charges may ultimately generate electric field spikes of the order of 1 GV/m. The fast response of ferroelectric materials to phase transitions under the influence of external electrical and/or mechanical stress allow us to contemplate the design of structures which emit and accelerate charged-particle beams.

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1. Introduction

Ferroelectric crystals (FEs) become spontaneously polarized so that the charge density on their end-faces can reach 10^{14} charges per square centimetre. Normally, these charges are compensated or screened by equal and opposite, external or internal surface charges. If a practical method can be found that will provoke a polarization change very quickly before charge compensation or screening can occur, thus leaving the surface charges exposed and in place, it will be possible to envisage the prospect of copious electron emission which might surpass that of present-day electron guns. The exposed surface charges are a potential source of accelerating fields (GV/m) which are tantalizing for the designers of the next generation of linear colliders such as the CERN Linear Collider (CLIC).

Since the ferroelectric properties of crystals are not generally well known to the accelerator community, we devote the bulk of this paper to a description of the phenomena, to the methods of inducing polarization reversal or phase transition either in a slow or prompt fashion, and to the possibility of triggering electron emission by laser illumination. First experimental results are reported and finally we permit ourselves to speculate on a few applications in accelerator technology.

2. Basic Ferroelectric Phenomena

2.1 Spontaneous polarization and screening processes

Ferroelectric materials exist as single crystals or as ceramics which, when cooled below their Curie temperature T_C , become spontaneously polarized. The high spontaneous polarization \vec{P}_s can be reversed by an external electric field. The process is characterized by a typical hysteresis loop. The spontaneous polarization is different from that induced in a normal dielectric by an applied field, since it is a permanent state of the crystal requiring no external field to sustain it. To polarize a normal dielectric to the same extent would require external fields of the order of GV/m.

At the Curie point, a ferroelectric crystal undergoes a phase transition as a result of collective ion sublattice displacement or permanent dipole moment orientation. The spontaneous polarization appears instantly at T_C (first-order phase transition, Fig. 1) for BaTiO_3 or continuously (second-order phase transition).

The perfectly aligned electric dipoles in an FE end at the opposite surfaces with unpaired positive and negative bound charges. Provided compensation by opposite charges can be prevented, the associated electric field in such a fully polarized system is given by $\vec{E} = -\vec{P}_s/(\epsilon_s\epsilon_0)$. In the case of PbTiO_3 , with $|\vec{P}_s| = 0.9 \text{ Cb/m}^2$, we expect an external field of the order of $100/\epsilon_r \text{ GV/m}$!

With an applied d.c. electric field E_{dc} an uncompensated polydomain structure can be generated, and even single-domain structures are formed. However, the enormous electric field strength due to \vec{P}_s will not build up to the theoretical limit and it cannot exist permanently. Neighbouring domains can align themselves in opposite

directions (180° structure), or can become distributed at 90° or 60° to each other depending on the type of FE. Various effects screen the spontaneous polarization and prevent the formation of a macroscopic

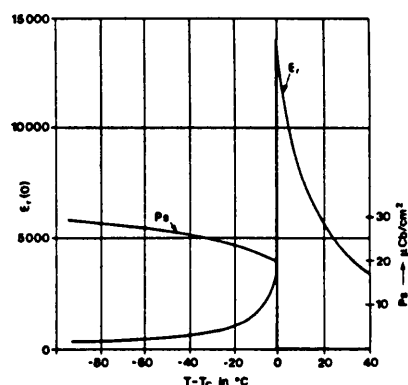


Fig. 1 Temperature dependence of spontaneous polarization \vec{P}_s and dielectric constant ϵ_r near the Curie point T_C , in case of a first-order phase transition (example BaTiO_3 single crystal).

field even though they do not cancel the spontaneous polarization of the crystal. Their principal causes are:

1. Electric discharges may occur through the surrounding medium between the surface of neighbouring domains of opposite polarization, and between such a domain and the electrodes. In single-domain structures, flash-over between different surfaces can take place.
2. Ion defects and electrons from the bulk of the crystal will screen \vec{P}_s in a time proportional to the resistivity. These free carriers build up an additional space-charge polarization $P_{sp.ch.}$, which plays a very important role in the dynamical behaviour of ferroelectrics. Whilst the free-charge carriers can be mutually neutralized, the bound surface charges due to \vec{P}_s can only be screened.

As a result of these screening effects, a dense distribution $P_{sp.ch.}$ builds up on the faces of the crystal, and at the same time the domains may be realigned. The surface charges screen the outside world from the spontaneous dipole moment of the crystal and the electric field associated with \vec{P}_s can appear only for a short time and generally with a lower amplitude than the theoretical amplitude.

In spite of the screening effects the main features of ferroelectrics—i.e. surface charge densities of as much as $10^{14}/\text{cm}^2$ and strong transient electric fields due to \vec{P}_s or $\vec{P}_{sp.ch.}$ —make these substances potentially interesting for the field of particle accelerators. However, these properties can be made to become visible in a real crystal only by choosing well-defined conditions, and by suppressing, as much as possible, the phenomena which work against their appearance. The purpose of this work is to point out possible ways to do this. The most promising conditions are expected during fast polarization reversal or fast phase transitions across the phase boundary, triggered externally by a fast electric field pulse and/or a fast pressure pulse.

2.2 Phase transitions by an electric field and mechanical pressure

The following phase transitions exist in FEs having not only paraelectric (P) and ferroelectric (F) phases (as in Fig. 1), but also an antiferroelectric (A) structure: $A \rightleftharpoons F \rightleftharpoons P$ (type I) and $F \rightleftharpoons A \rightleftharpoons P$ (type II). The phase boundaries are affected by temperature, pressure, and electric field. In antiferroelectrics (AFE) each neighbouring dipole chain is antiparallel-oriented, resulting in a zero macroscopic polarization. Nevertheless the crystal is in a microscopically polarized state. The PbZrO_3 crystals and solid solutions (PZT) with Ti content up to 5% are of the first type [1] (Fig. 2). The intermediate ferroelectric phase appears in a narrow temperature region below the Curie temperature ($T_C \equiv T_{FP}$).

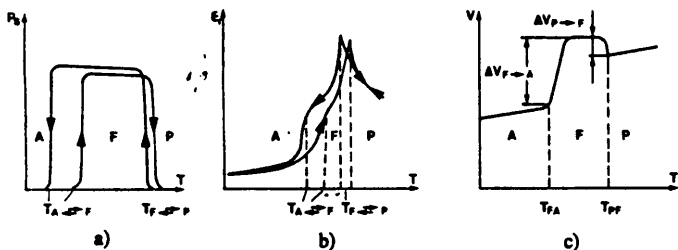


Fig. 2 The temperature dependence of spontaneous polarization P_s (a), electric permittivity ϵ_r (b), and unit cell volume V (c), for PbZrO_3 and $\text{Pb}(\text{Zr,Ti})\text{O}_3$ with A-F-P phase transition sequences [4, 5].

An applied electric field increases the temperature for the ferroelectric phase-transition $F \rightleftharpoons P$, whereas the temperature for $F \rightleftharpoons A$ is decreased in the case of the phase sequence A-F-P (Fig. 3a). Electric fields thus increase the width of the F-state [2, 3].

When mechanical pressure is applied, the result is opposite, giving rise to a narrower ferroelectric state (Fig. 3b). We should mention that the F-phase in those materials having a A-F-P phase sequence vanishes when the applied pressure exceeds a critical value ($p > p_c$) [2, 3].

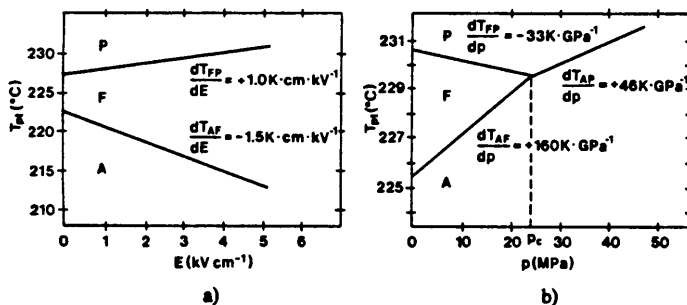


Fig. 3 The dependence of phase transition temperatures T_{pt} on the applied electric field strength (a) and on hydrostatic pressure (b) for PbZrO_3 [3].

Similar processes take place also in grains of polycrystalline (ceramic) materials. The macroscopic \vec{P}_s appears in such materials when an electric field is applied. Under the influence of the aligning field the resultant polarization of individual domains and grains is oriented according to the crystallographic conditions.

The $\vec{P}_{sp, ch}$ can be formed by an electric field as well by ion defects and free-electron displacement over macroscopic distances. Most of these defects are concentrated in a thin layer on the grain boundaries. Because of their increased mobility in these places, macroscopic $\vec{P}_{sp, ch}$ is formed on the surface layers of the ceramic samples. In the case of electroded samples, injected charges also play an important role. The $\vec{P}_{sp, ch}$ formed under a d.c. electric field at higher temperatures can be frozen by cooling under the influence of the field, forming a metastable poled state.

2.3 Diffuse phase transitions

The phase transitions in some macroscopic homogeneous materials are characterized by the fact that the phase-transition temperatures are not sharply defined as in classical FEs. These so-called diffuse phase transitions (DPTs) are spread over a certain temperature interval, resulting in a gradual change of physical properties in that region.

Local inhomogeneity in material composition and differences in internal electric field and mechanical strain cause the local phase-transition temperatures to differ in individual domains and grains. These internal factors have an influence on phase-transition temperatures which is similar to the influences of the external electric-field and the mechanical pressure described above. In individual domains the conditions for these transitions depend on the conditions prevailing in the vicinity of the domains. For the majority of domains, these phase transitions occur near the average phase-transition temperatures, whereas for a remarkable number of microdomains they take place at temperatures that are higher or lower. The macroscopically observed phase transitions are DPTs. Here, the difference in the electrical and chemical properties of the surface layers [4, 5] plays an important role.

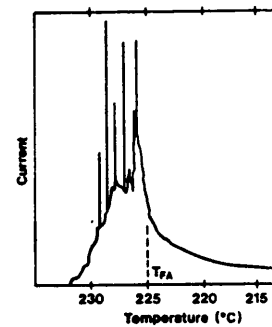


Fig. 4 Temperature dependence of pyroelectric current in PbZrO_3 during cooling. Strong Barkhausen pulses are observed on top of the normal pyroelectric current [5].

As an example, for the PbZrO_3 and $\text{Pb}(\text{Zr,Ti})\text{O}_3$ materials considered by us, the diffuse character of $F \rightleftharpoons P$ and $F \rightleftharpoons A$ phase transitions has been confirmed, based on X-ray and pyroelectric measurements [4, 5]. An example of temperature change in the pyrocurrent during cooling through $T_{F \rightarrow A}$ is shown in Fig. 4 [5]. A large number of Barkhausen pulses are visible on the background of the proper pyrocurrent.

3. Fast Polarization Changes

3.1 Electrical procedure

The switching time of the \vec{P}_s vector depends very much on the temperature and the applied d.c. electric field strength [6]. We consider now a prepoled sample having two phase transitions ($A \rightleftharpoons F \rightleftharpoons P$). The existence of space-charge layers at the surface of the crystal gives rise to a strong electric field directed perpendicularly to the surface. When we apply an electric field to the crystal in the direction opposite to its polarization, each existing antiparallel domain starts growing immediately by way of the existing domain-wall motion [7]. A fast change of \vec{P}_s under HV pulses will result in large surface charges (e.g. $0.1\text{--}1 \text{ C/m}^2$) and can be the source of short HV spikes as well as a source of electron emission.

One can make use of the phase-transition temperature shifts under HV pulses. However, in order to achieve a phase transition electrically, we must work at a temperature very near to the regions of the phase transition $A \rightleftharpoons F$ or $F \rightleftharpoons P$, as we are inside the A- or P-state (Fig. 3a). For a given HV strength, the sample will undergo a phase transition into the F-state, giving rise to the simultaneous appearance or disappearance of \vec{P}_s . The crossing of the phase boundaries can be as fast as nanoseconds. However, using a grid or

semitransparent electrode, we expect also strongly enhanced electron emission during quick phase transitions, in particular during $A \rightleftharpoons F$. It was previously shown [5] that in the narrow temperature region close to T_{AF} , there also exist compensated F-domains in the A-matrix. These permit a very rapid macroscopic \vec{P}_s forming and reversing speed, even in the subnanosecond range, because of the very fast sideways motion of existing domain walls without a need for nucleation of new domains. On the contrary, inside the F-state the \vec{P}_s change and reversal speed is controlled by the much slower process of nucleation of new domains with opposite polarization vectors.

3.2 Pressure procedure

Here we may have two cases: $p \geq p_c$ and $p < p_c$ (Fig. 3b). We again consider the prepoled sample at a temperature inside the F-phase. An applied mechanical pulse will give rise to the disappearance of the F-state. In such a case a HV spike, as well as electron emission due to the fast \vec{P}_s disappearance, is expected. The well-known electrical-mechanical transformation can be utilized to form fast and short mechanical pulses. The investigated sample, together with a piezoelectric elongator, forms a type of electrical-mechanical-electrical converter. Such converters allow the pressure to be changed very rapidly [8]. Here we should mention that the piezoelectric response has been reported to exist in at least a submicrosecond time scale [9], which makes this method very promising for fast polarization changes.

3.3 Optical-electrical procedure

The highest surface-charge densities are expected to be produced by the combined change of the \vec{P}_s vector and the liberation of space charges by fast laser illumination. This photoelectric effect will give rise to electron emission; hence FEs are potential candidates for replacing conventional photocathode materials. It is of particular interest to reverse the \vec{P}_s vector in such a direction that electrons are generated on the illuminated surface. In addition, this will help to increase the electron emission. However, in order to achieve this, the use of semitransparent electrodes is preferable.

4. Experimental Results

Electron emission by means of slow phase transitions (e.g. $F \rightleftharpoons P$) has been mentioned by several authors [10-12]. A TGS crystal was slowly (10 s) heated across the F-P phase boundary. With a β -spectrometer 10^8 - 10^9 electrons/cm² with 25 keV energy have been measured [13] in spite of only a second-order phase transition and of the low heating rate.

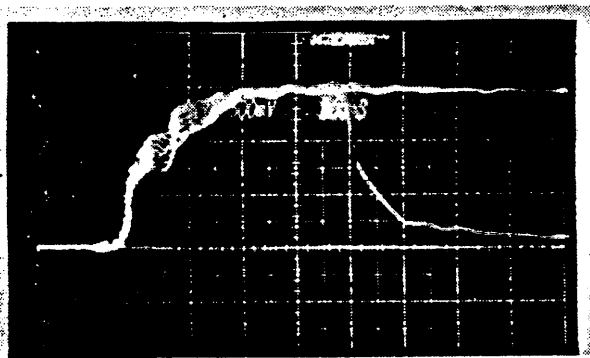


Fig. 5 Variation of HV pulses across a PZT (0.5% Ti) sample at 50 kV/cm amplitude during cooling from P-state 236 °C (upper traces) to the F-phase boundary 238 °C (lower trace with breakdown across sample surface). In the P-phase the pulse rise-time decreases with rising temperature.

Recently fast phase transitions ($\ll 100$ ns) have been performed with PZT samples by polarization reversal and phase transitions initiated by the HV-pulse method. Figure 5 shows the response of a

sample to many successive pulses at 50 kV/cm cooled down from 256 °C (P-phase) to the F-phase boundary. Breakdown across the sample occurred at 238 °C as a consequence of \vec{P}_s formation under electric field. The Curie point (under zero field) is $T_C = 233$ °C.

5. Applications in the Field of Accelerators

At this stage in the infancy of our understanding, the most hopeful application of ferroelectricity seems to be that of an electron-emitting surface capable of releasing some sizeable fraction of the 10^{15} charges/cm². Only 10^{10} electrons—provided they can be emitted in a short enough (< 30 ps) burst—would rival present photocathode surfaces and provide an electron gun for CLIC. Dramatic emission of electrons at the time of a phase transition is to be expected, and we can hope to control emission by illuminating the surface of a prepoled ferroelectric sample with pulsed laser light. Illumination with laser light ($\lambda < 1$ μ m) can cause liberation of electrons from the space-charge centres. Additional polarization reversal amplifies the electron emission. Our hope is not only to master this process but also to find an environment, parameters, and a geometry for the crystal, so that the large fields perpendicular to the surface layer can be used to give the electrons the initial acceleration they need in order to overcome their own space charge and produce a beam of small emittance.

Such ferroelectric photoemitting surfaces are not expected to be as sensitive to contamination as the materials used for present-day photocathodes, since they rely on the bulk properties of the crystals.

Another application which might stem from the laser-triggered or the polarization reversal modes is to make use of a short burst of electrons to trigger high-power switches.

High electric fields can only be obtained in an FE when the spontaneous polarization is changed rapidly. In order to be used in accelerator technology, the high fields must be precisely controlled in time. The electric field and the pressure methods are the most suitable since they offer fast and precise \vec{P}_s -reversal and transitions of the phase boundaries in both directions. In addition, the simultaneous illumination of certain ferroelectrics might well improve the density of surface charges. Devices based on these effects might be used as intense sources of electrons.

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