# *EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH*

/afm CERN PS/88-53 (AR)

## *FAST POLARIZATION CHANGES IN FERROELECTRICS*

## *AND THEIR APPLICATIONS IN ACCELERATORS*

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#### Abstract

Several mechanisms are described which change the polarization in ferroelectric material. Provided the change is too rapid for the related surface charges to be screened or neutralized, the high charge density can lead to strong electric fields. The fields may possibly be used for emission and acceleration of electrons. First results of experiments are reported, in which fast spontaneous polarization changes by reversal or phase transition have been demonstrated. Electrons of 25 keV energy have been observed, emitted from triglycine sulfate (TGS) crystals during phase transition while being slowly heated across the Curie temperature. With fast polarization changes, electron beams of even higher energy and density are expected.

> Paper to be submitted to Nuclear Instruments and Methods in Physics Research.

> > Geneva, Switzerland August 1988

## **1. INTRODUCTION**

Ferroelectric crystals become spontaneously polarized so that the charge density on their end-faces can reach 10<sup>14</sup> charges per square centimetre. Normally, these charges are compensated or screened by equal and opposite, external or internal, surface charges. If a practical method could be found that would provoke a polarization change very quickly before charge compensation or screening could occur, thus leaving the surface charges exposed and in place, it would be possible to envisage the prospect of copious electron emission which might surpass that from present-day electron guns. The exposed surface charges are a potential source of accelerating fields.

In this paper, experimental results are reported which demonstrate that a sufficiently rapid change in spontaneous polarization is possible, and that electrons of high energy can be emitted from ferroelectric surfaces during phase transition. So far, it has not been shown clearly that the surface-charge densities of  $10^{14}/\text{cm}^2$ , linked to spontaneous polarization changes, would represent fields approaching the order of GV/m, but we permit ourselves to speculate on some ways of achieving this.

Since the ferroelectric properties of crystals are not generally well known to the accelerator community, we describe the phenomena and the methods of inducing polarization reversal or phase transition either in a slow or prompt fashion, and we discuss also the possibility of triggering electron emission by laser illumination.

### **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  $P_s$  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 (displacement phase transition) or permanent dipole moment orientation (order-disorder phase transition). At  $T_c$ , the spontaneous polarization appears instantly (first-order phase transition) or continuously (second-order phase transition). A typical example of the temperature dependence of  $\overline{P}_s$  and  $\epsilon_r$  (dielectric constant) for a BaTiO<sub>3</sub> crystal having first-order phase transition is shown in Fig. 1. Above  $T_c$  in the paraelectric phase (P phase),  $\vec{P}_s$  is zero. At  $T_c$ ,  $\vec{P}_s$ jumps to 20  $\mu$ C/cm<sup>2</sup> and  $\epsilon$ <sub>r</sub> has a maximum of 14,000.

The perfectly aligned electric dipoles in a ferroelectric terminate 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  $\vec{E} = -\vec{P}_s/(\epsilon_r \epsilon_0)$ . In the case of PbTiO<sub>3</sub>, with  $|\vec{P}_s| = 0.9 \text{ C/m}^2$ , we expect an external field of the order of 100/ $\epsilon_r$  GV/m!

When a d.c. electric field E<sub>de</sub> is applied, an uncompensated polydomain structure can be generated, and even single-domain structures are formed. However, the enormous electric field strength due to  $\overline{P}_5$  will not build up to the theoretical limit, and it cannot exist permanently. Neighbouring domains can align themselves in opposite directions  $(180^\circ \text{ structure})$ , or can become distributed at  $90^{\circ}$  or  $60^{\circ}$  to each other, depending on the symmetry of the ferroelectric crystal structure. Various effects screen the spontaneous polarization and prevent the formation of a macroscopic field, even though they do not cancel the spontaneous polarization of the crystal. Their principal causes are as follows:

## *4.* **EXPERIMENTAL RESULTS**

Electron emission by means of slow phase transitions (e.g.  $F \rightleftarrows P$ ) has been claimed by several authors [10–12]. A triglycine sulfate  $(NH_2CH_2COOH)_3 \cdot H_2SO_4$  (TGS) single crystal was slowly heated across the F-P phase boundary over a time of <sup>10</sup> s. In spite of only a second-order phase transition and of the low heating rate (0.2°/s), between  $\beta$  spectrometer, 10<sup>5</sup> and 10<sup>6</sup> electrons per cm<sup>2</sup>, with 25 keV energy, have been measured with a  $\beta$  spectrometer (Fig. 5). The emission started in bursts, a few degrees below  $T_c$  and stopped completely above it. By taking into account the momentum resolution of the spectrometer, the total number of electrons (15-25) keV emitted from TGS during phase transition is  $\sim 10^8$ -10<sup>9</sup> per square centimetre. The TGS samples were never raised to the Curie temperature ( $T_c = 49 \degree C$ ) after their production, prior to the measurement. They had no electrodes, and the electric field during the experiment was zero.



Fig. 5 Pulse-height spectrum of electrons measured with a  $\beta$  spectrometer for two temperatures of a TGS crystal (5 mm  $\oslash$  and 3 mm thickness) during heating. Near T<sub>C</sub> the measured electron energy was  $25 \text{ keV(a)}$ . The PM background is shown in (b).

Fast phase transitions (< <sup>100</sup> ns) have been performed with PZT samples, using polarization reversal and phase transitions initiated by the HV-pulsc method. Figure 6 shows the response of a  $Pb(Zr_0,995, Ti_0,005)$  sample to many successive rectangular pulses of 50 kV/cm amplitude. The sample was cooled down from 256  $\degree$ C (P phase) to the F phase boundary while being continuously pulsed with positive polarity. Breakdown of voltage (drop in U the lower trace in Fig. 6) across the sample occurred at 238°C as a demonstration of  $\vec{P}_s$  formation under electric field (see Fig. 3). The Curie point (under zero field) is  $T_c = 233^{\circ}$ C. When going back to the P phase (> 238°C for 50 kV/cm), the normal rectangular wave form reappeared.



Fig. 6 Variation of the rise of the HV pulse wave-form shape across a PZT (0.5% Ti) sample at 50 kV/cm amplitude during cooling from the P state, 256  $^{\circ}$ C (upper traces), to the F phase boundary, 238  $\degree$ C (lower trace with breakdown across the sample's surface). In the P phase the pulse rise-time decreases with rising temperature.

## **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  $10^{14}$ charges per square centimetre. High-density electron emission of 25 keV has been shown with a very slow phase transition without any external electric field. Much denser and more energetic beams can be expected by using the fast phase transition or polarization-reversal methods. Only  $10^{10}$ electrons — provided they can be emitted in a short enough  $(< 30 \text{ ps})$  burst — would rival present photocathode surfaces for electron guns. Strong emission of electrons at the time of a phase transition is to be expected, and one might hope to control emission by illuminating the surface of a prepoled ferroelectric sample with pulsed laser light. Illumination with laser light ( $\lambda$  < 1  $\mu$ m) can cause the liberation of electrons from the space-charge centres. Additional polarization reversal may amplify 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 <sup>T</sup>hey need in order to overcome their own space charge and produce a beam of small emittance.

One may also hope that ferroelectric photoemitting surfaces will not be as scusilivc Io contamination as the materials used for present-day photocathodes, since they arc ceramics and they rely partly on the bulk properties of the materials. 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 a ferroelectric when the spontaneous polarization is changed rapidly. In order for to be used in accelerator technology, the high fields must be precisely controlled in time. The electric pulse techniques and 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.

## **Acknowledgements**

We wish to thank C. Günther and his co-workers B. Ackermann and V. Grafen of the Inst. für Strahlen- und Kernphysik of the University of Bonn for preparing and performing our measurements with the *β* spectrometer.

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