EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH
Laboratory for Particle Physics

Divisional Report **CERN LHC/97-10 (DLO)**

Displacement and Emission Currents from PLZT 8/65/35 and 4/95/5 Excited by a Negative Voltage Pulse at the Rear Electrode

G. Benedek*, I. Boscolo*, J. Handerek**, S. Marchesini*, C. De Martinis*, H.Riege*** and A. Scurati*

Abstract

It is shown that non-prepoled PLZT ceramics, both in ferroelectric and antiferroelectric phase, emit intense current bursts when a negative exciting voltage is applied to the rear surface of the cathode. The spontaneous polarization induced in the bulk by applying the field through the cathode disk, creates a sheet of negative charge on the diode boundary of the ferroelectric. This, in turn, induces such a high electric field at the diode dielectric surface that electrons are ejected out from the ceramic surface into the vacuum. The coherent behaviour of the displacement and emitted current shows clearly that the emission is due to a variation of spontaneous polarization. A second effect generated by the application of the high voltage pulse at the rear side is the formation of a surface plasma. Applying a positive voltage to the anode, electrons are readily transferred through the diode gap.

* University, INFN and INFM, Milan, Italy
** University of Silesia, Katowice, Poland
*** CERN, LHC Division

Nuclear Instruments and Methods in Physics Research A

Administrative Secretariat
LHC Division
CERN
CH - 1211 Geneva 23

Geneva, Switzerland
25 November 1997

1- Introduction

A cathode composed by a ferroelectric disk sandwiched by a uniform electrode at the rear surface (RE) and a gridded electrode at the diode side (DE), emits high current pulses when properly excited by a fast electric field. At CERN experiments [1-3], the exciting field was directed from the front emitting surface of the cathode disk to the rear surface (negative excitation), whilst it was opposite in other experiments (positive excitation) [4-7]. In a previous paper [8] we have presented some results for the case of positive excitation, in this paper we present experimental results and the relative discussion for the case of negative excitation. We have studied the behaviour with and without an accelerating field through the gap of the diode, with the aim of selecting the energetic electrons from the rest and so to single out the ferroelectric effect from the metal-dielectric discharge effect. The samples in our experiments where not prepoled, contrary to experiments of ref. [1-3]. The FE materials are made from Zirconium-rich Lead-Lanthanum-Zirconium-Titanate PLZT x/y/1-y, of composition $[Pb_{1-x},La_x][Zr_y,Ti_{1-y}]O_3$, where x and y are the atomic fractional compositions of *La* and *Zr* . This material is called in abbreviation PLZT. We have investigated PLZT of 8/65/35 and 4/95/5 type, that is with low and high Titanium content (we remark that in papers 1÷3 it was written 2/95/5 instead of 4/95/5, because there 2% was intended 2 mole percent of La_2O_3 which corresponds to 4 atom percentage). The two kinds of material have a different phase state at room temperature, as shown in Fig. 1. The 4/95/5 material makes an antiferro (AFE)-ferroelectric (FE) transition under the action of a high enough electric field, while the 8/65/35 makes simply a piece of hysteresis loop under the action of an electric field.

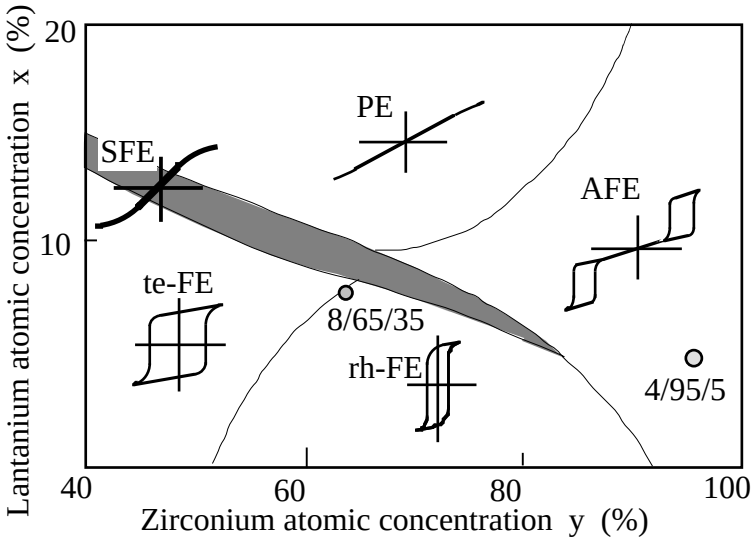

Fig.1 Room temperature phase diagram for PLZT materials showing ferro-electric (FE), antiferro-electric (AFE) and para-electric (PE) phases as function of composition. The SFE dashed region corresponds to a diffuse, metastable ferroelectric phases, that exhibits a gradual change of physical properties [from ref. 9]. PLZT 8/65/35 is in ferroelectric phase, while PLZT 4/95/5 is in antiferroelectric phase.

The peculiar characteristic of the ferroelectric emission from the ceramic is that electrons are energetic, contrary to thermoionic and field emission (and also photoelectric). Different physical mechanisms have been proposed to explain the ferroelectric emission. Here,

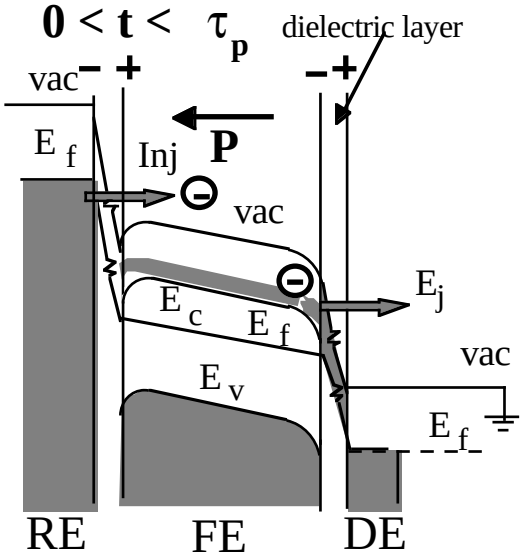

Fig. 2 Sketch of the band after the application of negative voltage pulse at RE electrode: Inj are electrons injected from the metal into the crystal surface region; vac is the crystal vacuum level, E_f, E_C and E_V are respectively the Fermi, the conduction bottom and the valence top levels, Ej are the electron ejected into the diode gap. We point out that there is a high jump within the dielectric layer.

following the line of previous paper [8], we try to give reason of it through the band diagram. The energy diagram for electrons in the ferroelectric crystal under the action of the negative field at the RE electrode, that can be figured out, is shown in Fig. 2. In PLZT crystal the high voltage pulse creates a spontaneous polarization P_S (pointing towards the RE electrode) with a typical charge density of 50 $\mu C/cm^2$ at the boundary of the ferroelectric disk, which, in turn, generates an electrostatic field of 10^6-10^8 V/cm on the surface dielectric layer (this layer has a thickness of about 1 μm) [10]. The high electric field induces a strong bending of the bands at the boundary of metal and vacuum. The electrons from the Fermi level of the metal are injected into the conduction band of the dielectric via tunnelling (the impurity *La* atoms provide the necessary donors centers for this tunnelling). The electrons are lifted above the vacuum level of the DE side (at the part of the surface not covered by the metal film). The electrons are acted upon by a so strong electric field that many of them exit the crystal surface with some keV. Obviously, part of the emitted electrons go to ground through the metallic film which covers the surface. The emitted electrons leave a layer of positive charges at the surface which screen the electron layer due to the spontaneous polarization.

We notice that electrons are injected into the rear side of the ferroelectric. They will screen the bound hole charges due to P_S.

2- Experimental Results and Discussion

The scheme of the experimental setup is shown in Fig. 3.

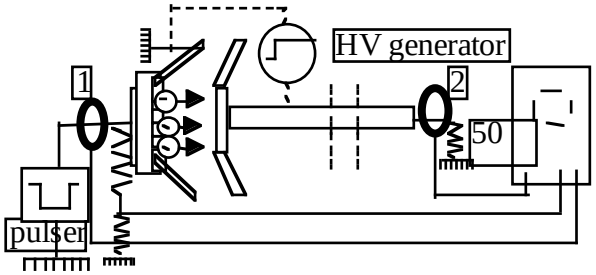

Fig. 3 Sketch of the setup for displacement and emission current measurement; the two currents are measured by Rogowski coils 1 and 2, a voltage divider measures the voltage pulse. The cathode is inserted in a Pierce designed diode. A decoupling capacitor is inserted when an accelerating voltage is applied.

The cathodes are disks having thickness less than 1 mm and diameter ϕ =16 mm. On the central area of the two sides a very thin layer of silver paste has been deposited for a diameter of $\Phi \approx 8$ mm on the DE surface and of $\Phi \approx 5$ mm on the RE surface. The emitting surface appeared under the optical microscope as "pock-marked", with many points of bare surface. The typical working pressure within the vacuum chamber was 10^{-4} mb. The HV pulser consists of a 10 nF capacitor connected to a parallel of two fast switches (6 ns rise time and 15 ns decay time) gated by a rectangular wave generator (HP 8114A) of features 1A-10 V- 150 ns.

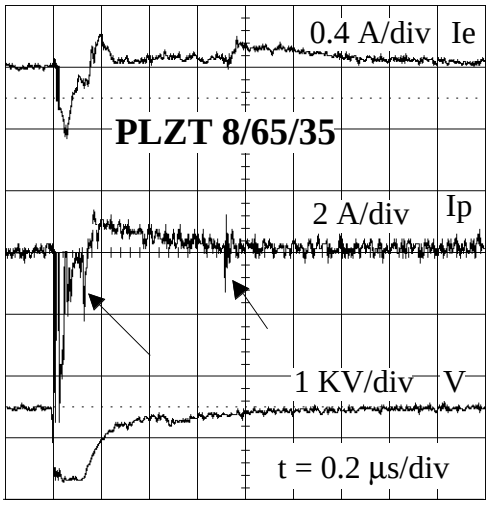

Fig. 4 Typical current waveform for PLZT 8/65/35: from the bottom, the first signal refers to the exciting voltage, the second refers to the displacement current and the third refers to the emitted current; the arrows indicate the displacement current peaks.

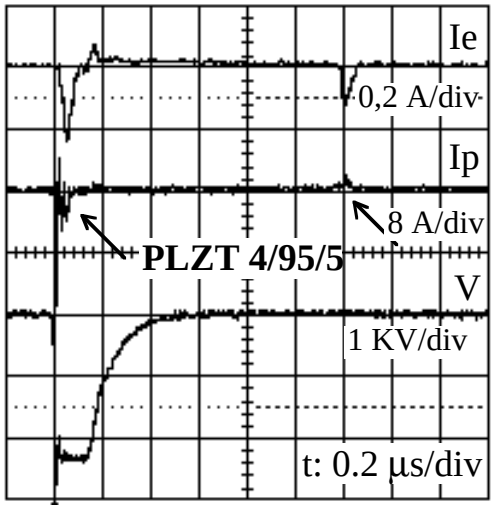

Fig. 5 As in fig.4 but for PLZT 4/95/65.

In Figs 4 and 5 typical current signals, obtained without accelerating voltage, are shown. They refer respectively to 8/65/35 and 4/95/5 samples. The threshold for electron emission comes out to be about two times higher for 4/95/5 than for 8/65/35. This is explained by the fact that the former material must make the AFE-FE transition (see Fig. 1) before doing the hysteresis loop.

The central signal in the two frames, recorded by Rogowski coil 1 in Fig. 3, is the current feeding the sample capacity during the pulse. It results from the superposition of two terms: $I = I_c + I_p$, I_c is the normal capacitor charging current ($I_C = C_\varepsilon \, dV/dt$), I_p is the non linear polarization current coming from the variation of the spontaneous polarization, $I_p \propto dP/dt$ (indicated by arrows in Figs 4 and 5). The variation of the spontaneous polarization entails a variation of the screening charge. From the signals we see that the peaks of the displacement and emission current show up coherently in time, as expected. The waveforms of I_p and I_e show that their width follows the width of the exciting pulse for the ferroelectric sample 8/65/35, whereas the two current widths are narrower for the

antiferroelectric sample 4/95/5. Furthermore, for the samples of this last type, we have seen that they always emit a second current I_e burst (and sometimes more than one) at the end of the exciting pulse (when the polarization relaxes back), while 8/65/35 samples emit that second peak once a while (in Fig. 4 is shown the latter case), and never a third peak was emitted.

The peaks following the first one cannot be explained by the picture given above. We can say that, in general, any external perturbation upsets a state of screening charge and the crystal, in going towards a new compensate charge state, supports surface polarization switching, at least locally, which push away electrons. In samples of 4/95/5 type, the variation of polarization ought to be so fast and strong, owing to the FE-AFE phase transition entailed by the relaxation of spontaneous polarization, that some polarization oscillations are likely to occur. We must also consider that, at the moment of the external electric field extinction, a plasma sheet covers the crystal surface, see below. Hence, a variation of the surface potential is promptly transferred in oscillations of the front free electrons.

The neat and narrow I_p current peak after the front edge of the voltage pulse for 4/95/5 material, shows that the AFE-FE phase transition driven by the applied voltage is fast. This gives reason of the short and neat I_e burst.

We point out that the ferroelectric sample 8/65/35 starts to change its spontaneous polarization during the leading edge of the exciting voltage pulse, as shown by the I_p current signal and the emitted current signal I_e as well. The polarization current has, instead, a delay with 4/95/5 material, as, obviously, the emitted current. This is due to the fact that this latter material is not ready for the hysteresis loop at the application of the electric field. This consideration is furtherly supported by the fact that the initial overshoot in the voltage pulse is cut only with 8/65/35 sample: this starts promptly to absorbs charge from the external circuit. The delay of 20-40 ns for the emission in 4/95/5 sample is the time required for the formation of FE nuclei and domain wall motion [3].

The first I_p and I_e peaks are stable within a 20% scatter, while the second peaks are unstable both in amplitude and in time: the stability of the first peak comes about because the emission is driven by the leading edge of the voltage pulse, the instability of the second peak comes about because the emission occurs for the relaxation of the spontaneous polarization. From the thermodynamic theory, the state of the system set by the application of the electric field is metastable.

Finally, we remark that the electrons of Figs 4 and 5 are energetic because they had to overcome the space charge barrier (from simulation, with e-gun computer program, an energy of few keV have been estimated).

The emission of electrons from the crystal surface as described above is not the only phenomenon occurring at the application of the negative high voltage pulse at the RE

electrode: there is also a surface discharge initiated from the metal-dielectric contact, with the formation either of a surface plasma [11], or of an electron cloud as suggested in ref. [5]. In the microgaps between the metal film and the ceramic surface, the electric field is very high, (a good emitting PLZT ceramic is porous) and a tangential component is always present. The electrons emitted by the dielectric whether for the band bending or for the extraction by the electric field via field emission are multiplied by secondary emission from the metal film and from the adsorbed gas layer [11].

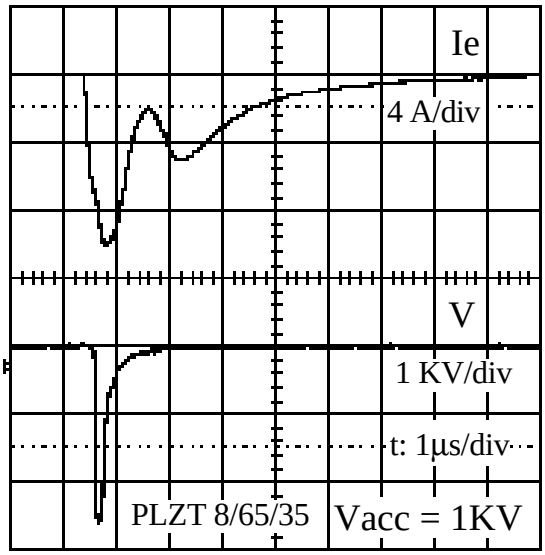

Fig. 6 Current waveform (upper trace) emitted by PLZT 8/65/35 when an accelerating voltage of 1 kV through the 1 cm gap is applied. The signal is the result of an average (made by oscilloscope) of 100 shots.

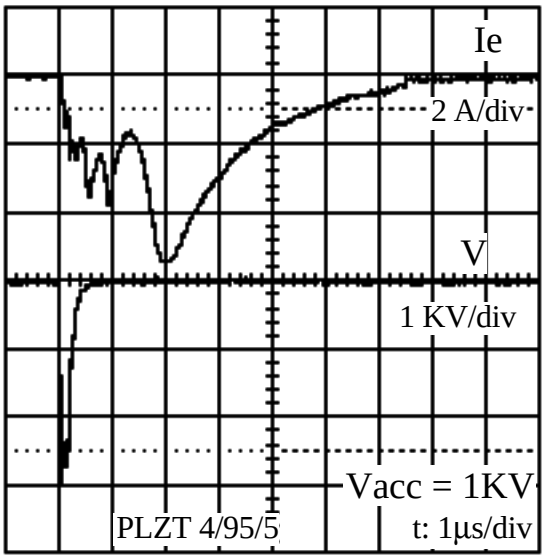

Fig 7 As in fig. 6 but with PLZT 4/95/5. Here the signal could not be averaged over many shots because they were too scattered.

The electrons emitted because of the surface discharge can be collected only by applying an accelerating voltage through the diode gap. In Figs 6 and 7 the currents obtained with the application of an accelerating field of 1 kV/cm are shown. The current amplitude jumps up from a fraction of Ampere to about I_e~8 A for 8/65/35 sample and I_e~4 A for 4/95/5. The impedance of the diode results less than 50 Ω. The high current together with the low value of the impedance lead to conclude that a plasma forms at the surface of the cathode (or anyway an electron cloud). We notice that the plasma does not close the diode gap, as observed with positive excitation (see [8]).

Both waveforms of the two signals show two peaks (the first peak of signal in Fig. 7 has superimposed an oscillation): they are related with the polarization and depolarization of the sample.

We notice that the "plasma" emission is lower in 4/95/5 ceramic with respect to the other type of material and, further, the onset of the current appears later.

3- Conclusions

Both PLZT 8/65/35 and 4/95/5 emit intense bursts of energetic electrons when excited by an electric field directed from the emitting surface towards the rear surface of the cathode disk. The 4/95/5 ceramic makes a very fast antiferro-ferroelectric phase transition (8/65/35 is already in ferroelectric phase). The first peak of emission current results stable within the 20% (and even better), while the other peaks show a larger scatter in intensity and time. This is due to the fact that in the first peak both the polarization change and the phase transition are driven by the HV pulse, while the other peaks occur because of the relaxation process. Incidentally, the reproducibility of the first current peak makes this configuration more suitable for electron sources than the configuration with the opposite versus of the exciting field, because in the latter case the emission peaks show a larger scatter.

We have shown that these cathodes do not need the prepoling (that is the setting of a spontaneous polarization and of a sheet of free carriers on the emitting surface) for the electron emission.

The intensity of the emitted current by the two materials is comparable. The current intensity is almost the same both with negative or positive voltage pulse on the rear surface. The repetition rate is higher with 4/95/5 material.

Finally, we point out that these cathodes are of easy preparing and handling. Their good behaviour as electron emitters also with the common silver paste as electrodes, makes these cathodes interesting for their robustness and strong emission.

Acknowledgements

We recognize the contribution of A. Muzzini in the design and realization of the experimental setup.

References

[1] H. Gundel, J. Handerek, H. Riege, E.J.N. Wilson and K. Zioutas, Ferroelectrics 100,1,1989.

[2] H. Gundel, J. Handerek, H. Riege, E.J.N. Wilson and K. Zioutas, Ferroelectrics 109,137,1990.

[3] H. Gundel, J. Handerek, H. Riege, J. Appl. Phys. 69,975,1991.

[4] J. Asano, T. Imai, M. Okuyama and Y. Hamakawa, Jpn., J. Appl. Phys. 31,3098,1992.

[5] J. D. Ivers, L. Schächer, J. A. Nation, G. S. Kerslick and R. Advani, J. Appl. Phys. 79, 2667, 1993.

[6] E. Zhang Conf. Proc. of the 1993 PAC, Washington D.C., May 1993.

[7] S. E. Sampayan, G. J. Caporaso, C. L. Holmes, E. J. Lauer, D. Prosnitz, D.O. Trimble, G.A. Westenskow, Nucl. Instr. Meth. Phys. Res. A 340, 90, 1994.

[8] G. Benedek et al. "Electron Emission from PLZT Ferroelectric/Antiferroelectric Cathodes Excited by Short High-voltage Pulses", submitted for publication.

[9] J. Handerek, J. Kwapulinski, M. Pawelczyk and Z. Ujma, Phase Transitions 6,35,1985.

[10] G. Rosenman, I. Rez, J. Appl. Phys. 73, 1904, 1993.

[11] V. F. Puchkarev, G. A. Mesyats, J. Appl. Phys. 78, 5633, 1995.