STABLE AMPERE LEVEL EMISSION OF ENERGETIC ELECTRONS BY ELECTRICALLY EXCITED FERROELECTRIC CERAMICS

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Abstract

Stable emission of energetic electrons from ferroelectric ceramic of PLZT type, i.e. lead zirconate titanate lanthanum doped, in form of thin disks have been obtained with a proper design of the front electrode. The current is about 1 A square centimeter and the energy is a couple of keV. High density, more than 50 A square centimeter, is obtained in the plasma assisted configuration. The electron expulsion from the surface is due to the spontaneous polarization switching induced by a high voltage pulse applied to proper metallic electrodes deposited as thin film on the two surfaces of the disk. In passing from the usual front electrode having the structure of an interconnected grating, to an electrode being a pattern of disconnected patches contained within a ring, the electron emission passes from erratic with samples and decaying with the number of shots to stable in both cases. This change in the behavior is explained by the fact that the sandwich of the continuous electrode and the grating constrains the domain switching and the relevant charge carriers motion within the part of material covered by metal, whilst a quasi-free surface allows the domain switching all over the whole area with the consequent flux of electrons in and out of the sample surface. In the plasma assisted emission, the front electrode must be the normal grating because the plasma in this electrode configuration is homogeneous.

1 INTRODUCTION

Electrons emitted from a ferroelectric cathode [1] come from ground to the crystal surface and then they are acted upon by the spontaneous polarization. This vector can be set in either direction with respect to the front (emitting) surface, depending on the sign of the voltage pulse applied at the rear electrode. With positive excitation (negative voltage), surface domains switch with the positive side pointing towards the front surface and then relax back. Therefore, in the first stage they attract screening electrons, then they push them out. With negative excitation, the negative side of the spontaneous polarization sweeps out the electrons loosely bound at the surface defects.

The process of excitation and relaxation must be of non-equilibrium type in order to have strong emission, so that the surface electron diffusion in the evolution towards an equilibrium state occurs through the material-vacuum interface [2]. Hence, the ceramics suitable for electron emission are the so-called ferroelectric relaxors [3] and antiferroelectric material with fast transition under the action of an electric field. The material used in the emission experiments was lead lanthanum zirconate titanate, called PLZT, with components proportion 4/95/5 and 8/65/35, where the numbers refer respectively to lanthanum, zirconium and titanium atom percentage. PLZT 4/95/5 material is in antiferroelectric (AFE) phase at the working room temperature, and it makes the transition to FE phase under the action of an electric field higher than 15 kV/cm, while PLZT 8/65/35 is in FE phase [4]. The emission observed [4, 5] with ceramic disks having the usual grating interconnected by a metallic ring, see fig.1, as front electrode had stable behavior neither with the number of shots in a single sample, nor in the level of emission (many times the emission was zero) for samples equal in composition, preparation and electroding. Instead, the emission obtained with an ensamble of metal islands surrounded by a metal ring was quite stable.

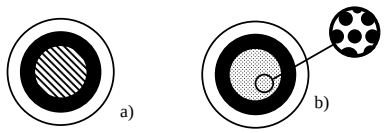

Figure 1: Sketch of the two electroding types of the front surface: the stripes are 200 μm with an interdistance of the same width.

The application of an electric field higher than 10 kV/cm through the samples, induces not only the spontaneous polarization switching within the crystal, but also the plasma formation on the cathode surface [4]. Both processes generate electrons, but the former process generates a group of electrons that are energetic, whilst the latter generates only non-energetic electrons.

We call the emission due to the switching of the spontaneous polarization, ferroelectric emission (FE).

2 DISCUSSION OF THE RESULTS WITH THE GRATING AS ELECTRODE

The set-up is described in ref [5]. A virgin emitting sample typically starts to emit FE electrons, then the emission slows down and finally stops after about one hundred shots. This cycle repeats after the rejuvenation of the sample. PLZT 4/95/5 samples show always switching peaks, because of the antiferro-ferroelectric transition, while non-emitting PLZT 8/65/35 samples show no sign of switching current after a certain number of shots. We have ob-

served that the ferroelectric emission is recovered, at least partially, after the reversion of the electrical pulse and after a long rest of the sample, one-two days.

The fact that switching and emission currents go to die in pair, lead to think that there is a mechanism which progressively blocks the domain switching of the uncovered stripes and/or confines the electric field under the metal stripes. This blocking mechanism could be due whether to a domain screening process, or to the formation of macrodomains so the spontaneous polarization does not relaxes back, as it occurs in normal ferroelectrics. The fact that many times there is no emission even if a well pronunced switching current is present (as is the case with PLZT 4/95/5) leads to say that either the switching occurs only inside the zones covered by metal, that is it does not propagate laterally, or the surface becomes passivated for screening and surface mutation due impurities, gas desorption, electron reduction, etc., or finally, the switching mechanism is not enough fast to prevent an evolution of the crystal configuration after the excitation through a complete internal migration of the charge carriers after multishots operation. The possibility of the confinement of the electric field under the metal area is supported by the growth of oxygen-deficient dendrite tips under the metal [6] which cause the increasing of the local field and the decreasing of the switching time.

3 RESULTS WITH A PATTERN OF METALLIC ISLANDS AS ELECTRODE

Different pattern of metallic islands have been tested: a) a pattern obtained by deposition of a uniform 100 Ågold film (this deposition auto-arranges itself in separate patches), b) a very thin silver paste film, which auto-arranges as a patchwork; c) a uniform filling of the surface holes (we remember that the material is porous) by a carbon paste and d) an Au evaporation with a mask of 50 μm diameter islands with an interdistance of 50 μm. A sketch of the pattern is shown in fig. 1.

The samples without the metal islands (with the external metal ring only) did not work.

We have observed that the samples emit always with the new type of electrode, but the nearer the interdistance between the islands the more stable the emission resulted. The results obtained by holes filling are reported.

No accelerating voltage has been set through the diode gap by purpose, because we wanted to catch at the anode Faraday cup only the energetic ferroelectric electrons. An accelerating voltage would have mixed the relatively few FE electrons within the huge amount of electrons extracted from the plasma sheet.

a) Negative pulse at the rear electrode

The signals of the emitted current reported in figs. 2,3 show a good stability. With PLZT 4/95/5 there are two current peaks, while there is only one for PLZT 8/65/35 and in this second case the signal amplitude is 2-3 times higher and longer than the first. These characteristics were

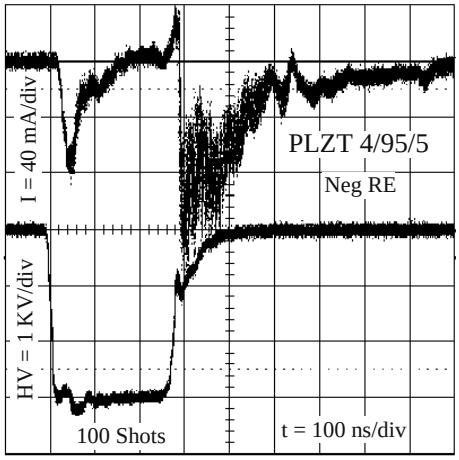

Figure 2: The superposition of 100 current signals, upper trace, and voltage signals, lower trace, is reported. The voltage pulse is applied to the rear electrode (RE), and the sample was a PLZT 4/95/5, as written in the frame, of 0.7 mm thickness.

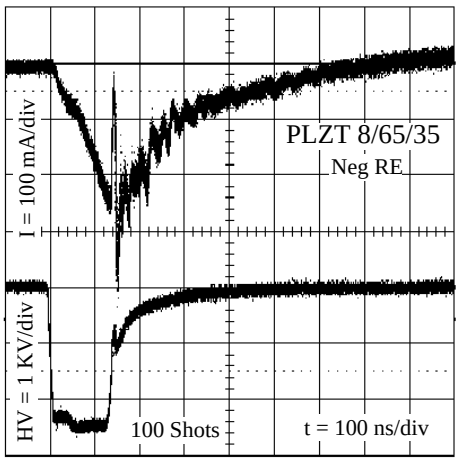

Figure 3: Ibidem as previous figure, but here the sample was a PLZT 8/65/35 of 1 mm thickness.

observed also with the grating, but here the second pulse in 4/95/5 material is substantially stable in time. For all the comparisons the reader is referred to the signals published in references [4, 5, 7]. The almost linear increasing of the 8/65/35 signal is explained by the continuous spontaneous polarization switching all along the excitation pulse. We notice that the very narrow pulses superimposed on the signals resemble the well known Barkausen pulses.

The first pulse is due to the building up of the spontaneous polarization oriented towards the rear electrode [5], while the second peak is due to the relaxation of the spontaneous polarization. The relaxation process of the spontaneous polarization is longer than its building up because in this latter case it is driven by the fast voltage pulse, and in the former case the ferroelectric state is metastable.

b) Positive pulse at the rear electrode

The signals of the emission are shown in fig.4 and 5.

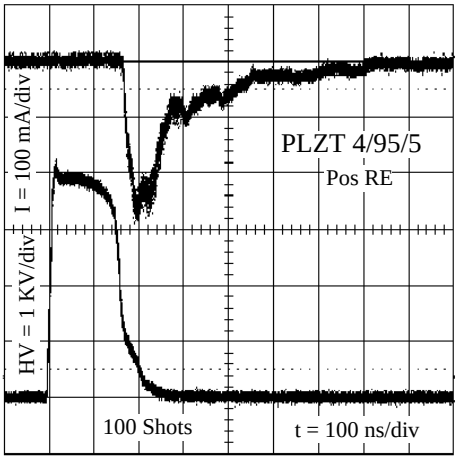

Figure 4: As in fig. 2 but with positive pulse to the rear electrode of a PLZT 4/95/5 sample.

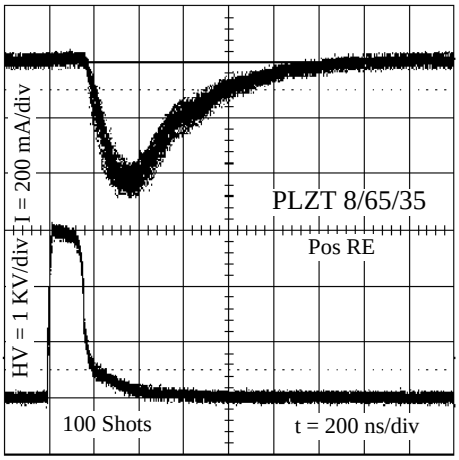

Figure 5: As fig.4 but for PLZT 8/65/35.

In this case the emission occurs at the relaxation of the spontaneous polarization P_s.

For the samples electroded with 100 Åfilm, the emission stopped after a certain number of shots because of the sweeping out of the metal islands.

4 DISCUSSION

The observations on samples with the grating lead to claim that a fast aging process is present for the application of a sequence of unipolar pulses. The neat separation of the cathode material in two parts, one under the metal stripes and the other one outside should induce a process of switching limitation within the covered zones and passivation of the uncovered surface.

The plasma which forms on the surface, owing to the high voltage of the excitation pulse, pushes further towards the surface passivation.

The screening and passivation processes do not occur for samples having disconnected metal patches as electrode. The disconnected electrode avoids the possible segregation of the electric field and so of the polarization switching under the covered zones. Our phenomenological view of operation is that the front of the voltage pulse propagates as a wave to the front surface, so inducing the co-propagation of polarization switching. As a consequence the surface switches into a substantial conduction state: in fact, the dP/dt current signal results in this configuration as high as with a completely electroded surface, further, in ref. [8] it was measured a high mobility of the charge carriers within the stressed region.

When the voltage excitation pulse is over, the surface goes to equilibrium by diffusion of the charge carriers through the surface towards the external ring or through the bulk. The paths of the electrons through the surface are not fixed because the surface state of a ferroelectric ceramic is dynamical and the surface is like a patchwork of peaces, whose properties [9] range from metallic to insulating.

We notice that in the grating case the plasma starts along all the rims of the stripes and covers the 200 μm in few hundreds of nanoseconds, in the other case that time is much longer because the plasma starts from the rim of the outside metal ring (it is connected to the pulser) and has to cover some millimeters of surface space.

It must be recognized that we are not able to give a clear phenomenological model explaining both the substantial unreliable operation with the first electroding and the reliable operation with this second electroding.

5 CONCLUSIONS

A stable copious emission of energetic ferroelectric electrons from a ferroelectric ceramic disk, under the application of a fast high voltage pulse, has been obtained when the electrode of the emitting surface was made as a uniform pattern of un-connected metal patches contained within a metallic ring.

6 REFERENCES

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