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FERROELECTRICS AND THEIR POSSIBLE USE IN ACCELERATOR PHYSICS

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ABSTRACT

Ferroelectric substances are characterized by the strongest spontaneous electric polarizations, being equivalent to $\approx 10^{15}/cm^2$ bound surface charges in some crystals. The associated macroscopic transient electric fields can be as much as 100 GV/m. Owing to the very limited lifetime of such field strengths, the main experimental task of this research activity is to establish them; some possible experimental techniques are considered. Practical arrangements to utilize these substances for charged-particle acceleration and beam bending are explained. A previous channelling experiment confirmed still much higher microscopic field gradients. The dynamical behaviour of ferroelectrics is explained, including our own experimental results (light emission and potential development across the crystal during switching). Switching times below microseconds have been achieved. A fast polarization reversal increases the probability of finding an uncompensated polarized state.

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FERROELECTRICS AND THEIR POSSIBLE USE IN ACCELERATOR PHYSICS

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ABSTRACT

Ferroelectric substances are characterized by the strongest spontaneous electric polarizations, being equivalent to $\approx 10^{15}/cm^2$ bound surface charges in some crystals. The associated macroscopic transient electric fields can be as much as 100 GV/m. Owing to the very limited lifetime of such field strengths, the main experimental task of this research activity is to establish them; some possible experimental techniques are considered. Practical arrangements to utilize these substances for charged-particle acceleration and beam bending are explained. A previous channelling experiment confirmed still much higher microscopic field gradients. The dynamical behaviour of ferroelectrics is explained, including our own experimental results (light emission and potential development across the crystal during switching). Switching times below microseconds have been achieved. A fast polarization reversal increases the probability of finding an uncompensated polarized state.

1. INTRODUCTION

Ferroelectric (FE) crystals are a special class of dielectrics (DE) [1]. Any other material inside the electric field gives rise to polarization phenomena, which last as long as the externally applied field. Ferroelectrics are the only materials that have a spontaneous and permanent macroscopic polarization $\vec{P}_s$, even in the absence of an electric field. The $\vec{P}_s$ is expressed as surface charge density (C/m^2), and bound charges are the origin of ferroelectricity. Each unit cell of the crystal has a resulting electric dipole. Many neighbouring cells can be aligned coherently, resulting in a macroscopic electric polarization, below the Curie temperature T_c, and giving rise to the appearance of domains. The terminology used for ferromagnetic materials (FMs) can be transferred completely if the magnetic properties are replaced by the corresponding electric ones. There is a simple difference between FEs and FMs: owing to the absence of magnetic monopoles, the magnetic field associated with an FM can exist permanently, whilst in FEs the resulting strongest electric fields are suppressed by

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external charges. In other words: the free external charges -- partly those on the crystal surfaces, partly those having migrated to the inside of the bulk of the material -- create an opposite electric field, diminishing the field $\vec{E}_s$ associated with the spontaneous polarization. However, the polarization state itself is not affected by the presence of these free charges. At this point it must be stressed that the crystal lattice in an FE below T_c is in an equilibrium state. The crystal environment is the one which does not accept the huge electric field gradients originating from such enormous electric dipole strengths, resulting in electrical breakdowns which are the origin of the external neutralizing charges mentioned above.

According to these considerations we must distinguish an FE state according to the time which has elapsed since its creation. When the neutralization process is finished, we are in the final and permanent state. In the beginning, during the transient time t^* we have to deal only with a pure polarized piece of matter. This is the time period in which we are interested here and where the strongest electric fields must exist. Therefore, the manifestation of the spontaneous polarization $\vec{P}_s$ is achieved *only* while changing the polarization, by varying either the temperature or the polarization vector direction. The latter is performed by applying an electric field. This is also the definition of ferroelectricity: we can change the polarization of a crystal from one state to the other by means of an external electric field.

In this type of research the main task is to investigate first the dynamical behaviour of these materials during the time interval t^* in order to choose the conditions that will keep t^* within reasonable values ($\approx$ps-μs). This is important for the detection of the electric fields as well as for their potential use in particle acceleration. It should be noted that this transient time t^* is different from the switching time t_s which is needed in order to perform a complete polarization reversal, although t^* can be much bigger than t_s. In a simplified picture, we might say that more time is needed to create a state than to destroy it. During a polarization reversal, we have a complete rearrangement of each individual dipole and, in addition, of the free external charges in the FEs. This fact must be taken into account in order to understand their dynamical behaviour and their response to external electric fields.

Finally, we must keep in mind that the coherent addition of the microscopic field of each electric dipole results in a macroscopic field. Although the surface charge formalism describes ferroelectricity to a large extent, it fails on a microscopic scale, and this is of the utmost importance for a deep understanding of these materials. Owing to the $1/r^3$ dependence of the field strength of an electric dipole, the microscopic fluctuations in an atomic scale are enormous. In this context we must mention antiferroelectrics (AFEs) which are also of interest in this work.

Because of the antiparallel arrangement of each individual string of perfectly aligned electric dipoles, they do not have any macroscopic polarization.

2. ELECTROSTATICS

The electric field inside and outside polarized matter is described by the polarization vector $\vec{P}_s$ [2]. Figure 1b shows the field distribution along the direction of polarization of a polarized disk, which is given by [2]:

$$\vec{E}_{in} = -2\pi\vec{P}_s \left[1 - \frac{\ell - z}{\sqrt{(\ell - z)^2 + a^2}}\right] -2\pi\vec{P}_s \left[1 - \frac{\ell + z}{\sqrt{(\ell + z)^2 + a^2}}\right] , \quad (1)$$

$$\vec{E}_{out} = 2\pi\vec{P}_s \left[\frac{z + \ell}{\sqrt{(z + \ell)^2 + a^2}} - \frac{z - \ell}{\sqrt{(z - \ell)^2 + a^2}}\right] . \quad (2)$$

For practical purposes the following relation is given:

$$1 \ C/m^2 \ \hat{=} \ 100 \ GV/m \quad (3)$$
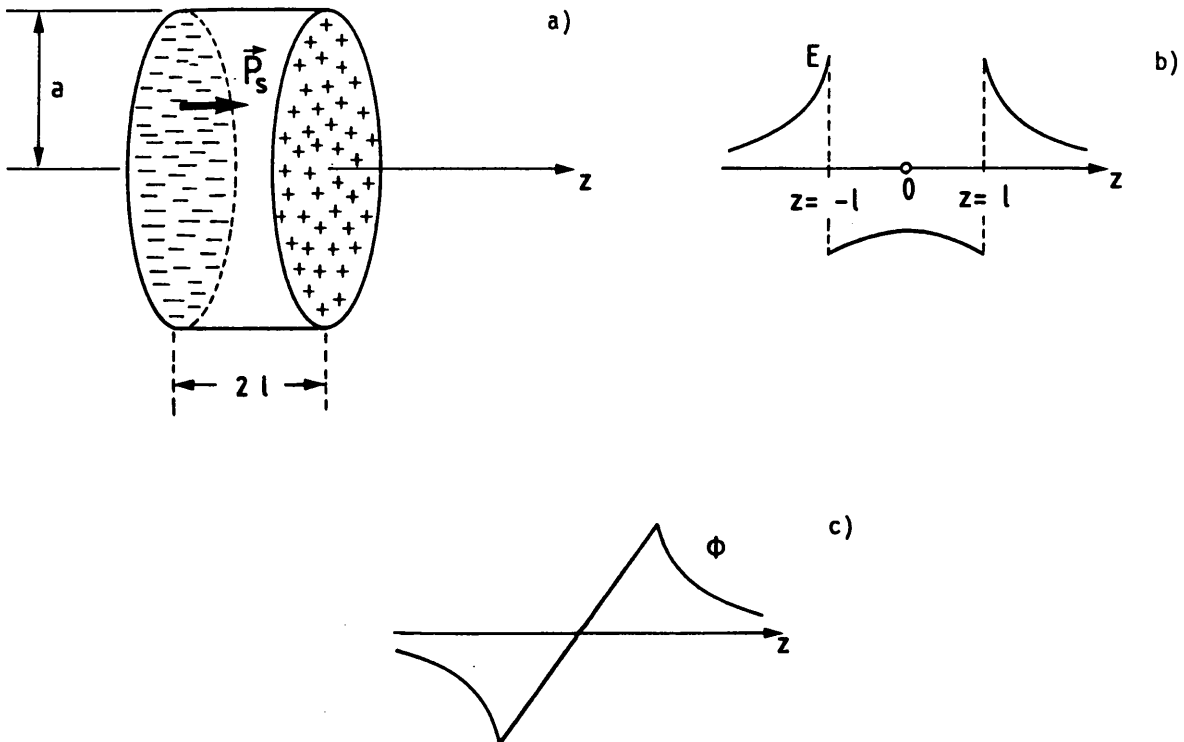
$[P_s(LiNbO_3) = 0.75 \ C/m^2]$.

Fig. 1 A cylinder uniformly polarized along its axis is described by bound surface charges a). The electric field b) and potential c) are shown.

This must be considered as a mean value. A fully polarized crystal is equivalent to a capacitor of the same dimensions, with charges $Q = \pm P_s$ on each surface; the inside is taken to be empty space. However, we have to consider bound charges. The energy needed in order to change the polarization of an FE substance from one direction to the opposite one is

$$W = -2\vec{P}_s \cdot \vec{E}_c \tag{4}$$

(E_c = coercive field).

In the above relations the dielectric constant ε of the material does not appear. This demonstrates the difference between the free charges on the surface of a DE being necessary for creating such a field, and the bound surface charges associated with the permanent polarization of the bulk of the crystal in the case of FEs. In the first case the presence of a free charge polarizes the surrounding material in such a way as to oppose its electric field (this being described by ε). In FEs the sequence is just the opposite: we have first the creation of the spontaneous polarization, which gives rise to the appearance of an electric field described by the bound surface charges. It must be mentioned that the spontaneous polarization of FEs is the strongest one known so far. An external electric field applied to a polarized FE crystal cannot polarize it any further. This being so, it means nothing more than that the FE is behaving as having $\varepsilon = 1$ once saturation has been reached. All these considerations apply, of course, to the transient time period t^*. During the steady case, we are again at the other extreme. The external neutralizing charges are loosely bound at the surface or inside the crystal. By applying an electric field externally, these quasi-free charges are very easily moved and therefore high values of ε are observed in the FE. These considerations are important for making the distinction between the transient period t^* and the permanent case of an FE.

3. ACCELERATION--BENDING OF CHARGED PARTICLES

Once the electric fields described above have been established, the bending efficiency on energetic charges of an FE is obvious if we keep in mind the capacitor-analog. However, according to the potential distribution of a finite (!) FE shown in Fig. 1c, no energy gain can be achieved if a charged particle traverses such a crystal. Nevertheless there are some practical arrangements which help to overcome such major problems and to utilize these electric fields also for particle acceleration. We mention the following:

i) Keeping in mind Fig. 1, we start with a negatively charged ion (e.g. H^-). Once it enters the crystal it will almost immediately become positively charged (p). The overall energy balance is now such that we might achieve particle acceleration or deceleration.

ii) If a charge is created inside the crystal, this again might well give rise to an overall energy balance that is different from zero. In another set-up, proposed by H. Riege, an electron bunch traversing the crystal through a small hole, while the crystal is being polarized, will interact with the transient electric field, giving rise to accelerated short electron bunches. In addition, if the electron bunch lasts as long as the switching procedure, some charges in the hole will always be synchronized with the transient time.

iii) Acceleration and simultaneous bending can also be achieved if the velocity of the particle is transverse to the electric field $(\vec{v} \perp \vec{E}_s)$. The energy gain is given by [2]

$$\gamma m = \gamma_0 m \sqrt{1 + \left[\frac{qE_s \; (GV/cm)}{\gamma_0 mc^2 \; (GeV)}\right]^2} \times \ell \; (cm) \qquad (5)$$

(q is the charge of the particle and ℓ is the path length inside the field $\vec{E}_s$).

A proton at 10 GeV or 1 TeV inside an electric field E_s = 1 GV/cm will be accelerated by 50 MeV/cm and 0.5 MeV/cm, respectively. A uranium nucleus at 5 TeV will still gain ≈ 1 GeV/cm, which is equivalent to a 10 MV/cm longitudinal electric field. Cylindrical-type arrangements, having also a hole of small diameter d (d $\lesssim$ crystal thickness!), seem to be promising. In such a case the crystal does not need to be very thick, and we avoid the scattering phenomena inside matter. These facts are of practical importance. In addition, we will have a tremendous bending force since:

$$300 \; T \doteq 100 \; GV/m \; . \qquad (6)$$

iv) More sophisticated techniques may be used for acceleration. The channelling phenomenon inside FEs and AFEs [1, 3] is mentioned. The observation made by Gemmell and Mikkelson (Fig. 2) [4] is shown already at this point: the channelling effect for 3.8 MeV protons in transmission in the {100} plane of $BaTiO_3$ diminishes when $\vec{P}_s$ is perpendicular to the planes and can best be understood in terms of the extremely high electric fields (~ 10^{10} V/cm) at right angles to the channel. We must bear in mind that the expected maximum field is much less: 250 MV/cm. Another feature of interest is the observed increase, by a factor of 2, of the angular width of the peak for particles in transmission in the ferroelectric state (0.12°) in comparison with the paraelectric state (0.06°). However, the immense importance of this measurement is due to the fact that the experiment was performed during the steady state. A deeper understanding of such behaviour seems to be of the utmost importance. This experimental result is the strongest evidence supporting this research activity.
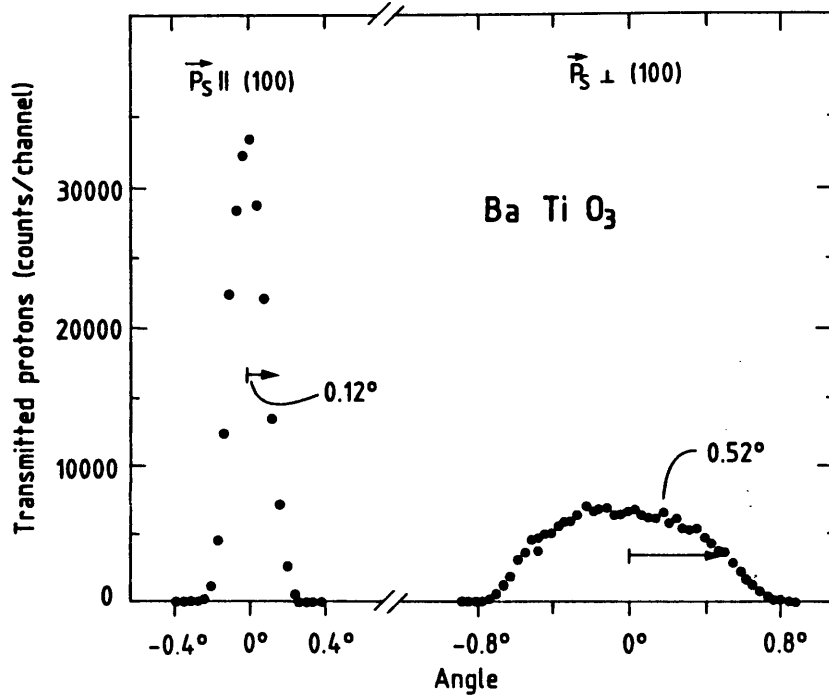
Fig. 2 Angular scan over the proton beam channelled along the (100) planes of BaTiO₃ with its polarization vector $\vec{P}_s$ polarized either parallel (left) or perpendicular (right) to the (100) planes [4].

4. DYNAMICAL BEHAVIOUR

In the previous section, we have described by electrostatics a dynamical system such as a freshly created polarized FE state. Nevertheless, this formalism is correct until the neutralization procedure becomes significant. Table 1 gives the threshold voltages for electrical breakdowns under different conditions [5]. The corresponding breakdown time intervals must be chosen to be maximum, or at least longer than the switching time t_s, otherwise the spontaneous polarization $\vec{P}_s$ will not reach its highest value -- the polarization charges will flow away whilst they are created. Therefore, the circuit used for the aligning voltage must have a small impedance while the voltage is being applied; this must become very large immediately after the switching process is terminated. In order to achieve a crystal polarization as high as possible and also a longer duration.

All the above requirements are satisfied by using a pulsed electron gun, as was proposed by H.J. Besch and T. Ypsilantis, in order to create the coercive electric field by charging the crystal surfaces. Ohmic contacts are avoided, so that the external charges can reach the crystal slowly. For this purpose, a pseudo spark chamber (PSC) [6], giving some 10^{12} electrons within ≈ 10 ns with 30-60 keV kinetic energy, seems to be a very attractive device. Polarization reversal times in the nanosecond region seem to be feasible

Table 1

Breakdown thresholds V_{th} in gasses at 1 atm [*]

Gas	Pulse duration	V_{th} (MV/cm)	Relevant polarized FE
Air	20 ps 3 ns	170 > 8	TGS $BaTiO_3$
N_2	7 ps	~ 870	$LiNbO_3$ $PbTiO_3$
H_2	200 ns	2	$K_4Fe(CN)_6 \cdot 3H_2O$

[*] According to [5] and to references [10-13]
therein.

according to our present experimental results. The basic idea behind the circuit proposed by Camlibel [7] is a convenient direct method for measuring the polarization charges appearing during the switching.

5. CRYSTAL LIFETIME

There are some 700 FEs [1]. Most of them cannot survive many polari zation reversals. However, there are quite a number of crystals that are very resistant to the associated mechanical stresses. Here we mention only those crystals with which we have already had experience within the requirements of this work: TGS, KFCT, $KNbO_3$, PZT, PLZT. These crystals are particularly convenient for studying the dynamical behaviour. The poly-crystalline types of PZT and PLZT combine simultaneously many properties that are important for our purposes: the high spontaneous polarization ($P_s \approx 0.5$ C/m^2) can be switched with 2-3 kV/mm only; fatigue effects appear slowly; they are not expensive; and they are machinable materials, widely used in advanced technology [8].

6. EXPERIMENTAL APPROACHES

The most convincing way to detect the expected electric field gradients directly is through their Coulomb interaction with a charged particle. Most of the experiments under way at CERN are mentioned below.

1) Potential measurements of the crystal electrodes[a]. In this systematic study of the behaviour of FEs during polarization reversal, we use an indirect method in order to measure the time-dependent potential at one

of the crystal electrodes without having any ohmic contact with the FE itself, and which might destroy potential spikes during polarization reversal. A pulsed e^- beam from a PSC is used (Fig. 3). If the potential is changing owing to the switching mechanism, even for sub-nanosecond time intervals, we will see a change in the upper limit of the associated bremsstrahlung spectrum. The sensitivity is at least in the kilovolt range. Finally, it must be mentioned that the same set-up can be used for polarization reversal. This being successful, and keeping in mind the enormous e^- intensity within ≈ 10 ns, we can combine fast polarization reversal and simultaneous potential measurement of an FE which is almost suspended in space.
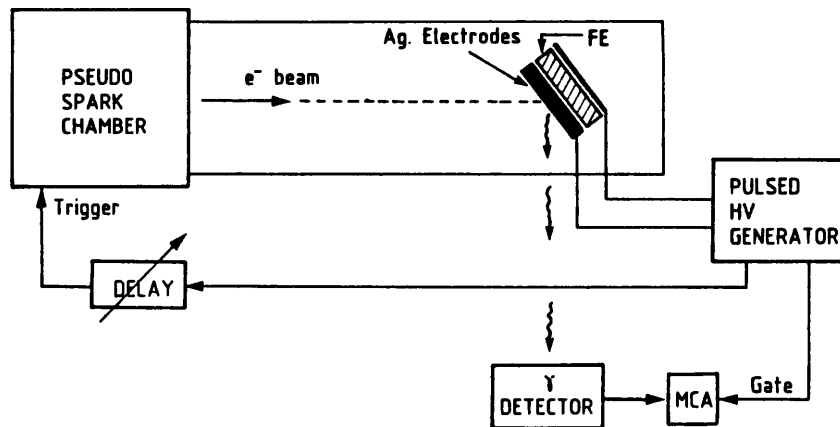
Fig. 3   Potential developments on the crystal electrodes during switching, studied by the electron beam from a PSC.

2) Creation of short accelerated electron bunches[a]. Figure 4 shows a simplified schema of this idea. An electron bunch from a PSC traverses the FE through the small hole. While the crystal polarization is switched on in one or the other direction and a ≈ 10 ns long e^- bunch is synchronously created, only a small part (≈ a few ‰ for crystals of < 1 cm) will be able to reach the other side of the crystal and, in addition, gain energy [see Section 2, point (ii)]. The t_s, t^*, and $\vec{P}_s$ will define the compression of the original bunch, the final energy gain, and the energy spread. Keeping in mind Table 1, we might well be able to change t^* through the surrounding conditions. Therefore the electrostatic relations will, in addition, change with time, resulting in a further selection on the e^- bunch, as far as the bunch compression and the energy balance is concerned. This will be possible, if the transient time t^* is of the order of the time needed for the charges to traverse the crystal.

3) Particle acceleration may be achieved by creating a charge inside the crystal. A pulsed K X-ray source[b] could be used for that purpose (Fig. 5). A pulsed hydrogen lamp illuminates the photocathode, giving
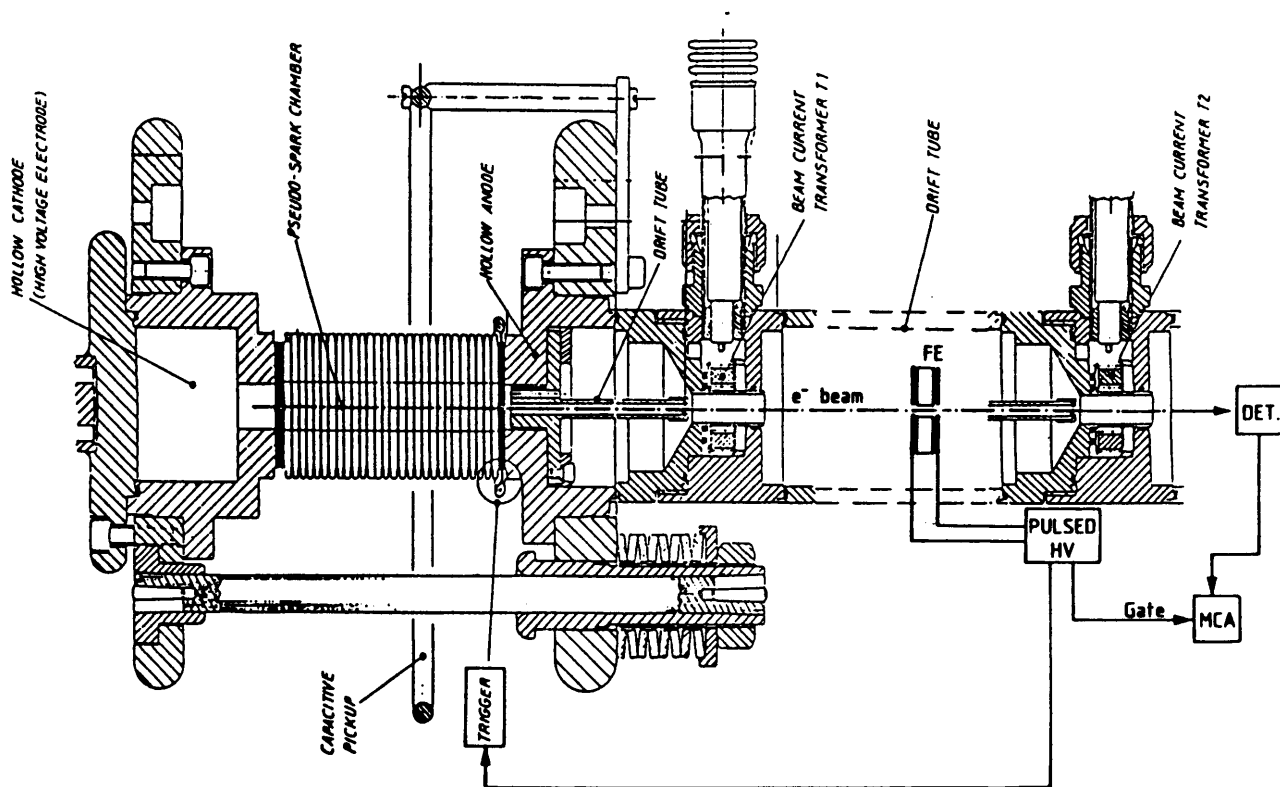
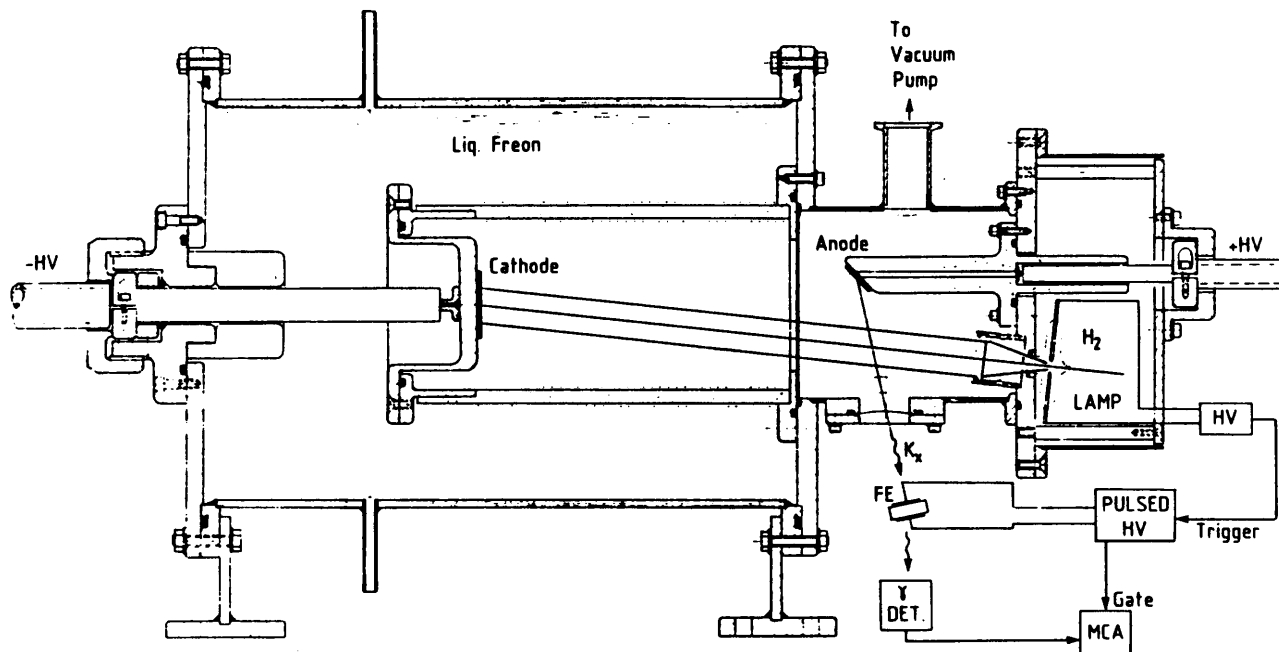Fig. 4   A pseudo spark chamber [6] used for measurements with FE.

Fig. 5 Schema of a pulsed X-ray source. The electronic circuit and the detection system are shown.

rise to electron bunches of a few nanoseconds (without a d.c. contribu-
tion), which will create K X-rays or bremsstrahlung while hitting the
anode. The maximum photon energy is at 100 keV and their yield is
~ 10^4 γ's/pulse. Their intensity and time structure can be changed
externally, through the lamp power supply. The interaction of these
photons via the Compton or photoelectric effect inside the bulk of the
crystal will give rise to secondary electrons. As was explained above,
such electrons will be finally accelerated provided this is not prohibi-
ted by unforeseen threshold effects.

4) Beam bending[b] seems to be a very sensitive detection technique. If the
spontaneous polarization $\vec{P}_s$ is transverse to the beam direction, the
field lines of the FEs are such that, inside and outside the crystals,
the bending force has always the same direction. The sensitivity of such
an experiment is increased enormously if a small hole is used inside the
crystal since the scattering effects are then avoided.

7.   PRESENT EXPERIMENTAL STATUS

7.1 Light emission

The light emitted during polarization reversal is correlated [9] to the
huge surface charges expected in any newly created FE state, as is demon-
strated in Fig. 6. We must keep in mind that the light emission is sup-
pressed after some few HV pulses, of the same polarity, applied to the
crystal. The remaining intensity is due to the rest-polarization of the
crystal. No matter whether the emitted light is due to discharges outside
the crystal or to electron acceleration inside it [9], this effect is
probably the best way of demonstrating the appearance of additional surface
charges/potential, excluding the applied HV pulse as their origin. In a
systematical study, the light emission distribution (Fig. 7) was confined to
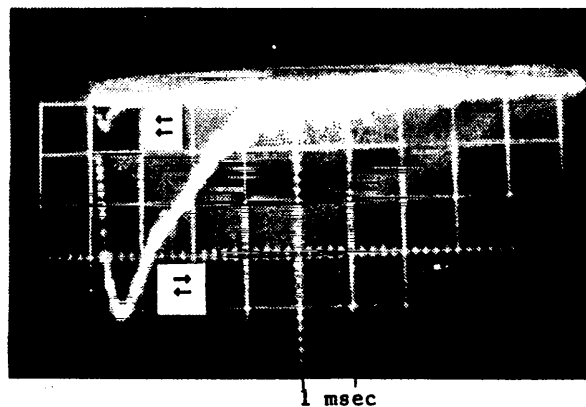~ 30 μs (FWHM), although each individual light-pulse was some 100 ns wide[b].

1 msec

Fig. 6 Light emission of a TGS crystal (0.5 cm^2 x 4 mm):   a) $\vec{\rightleftarrows}$ alternating
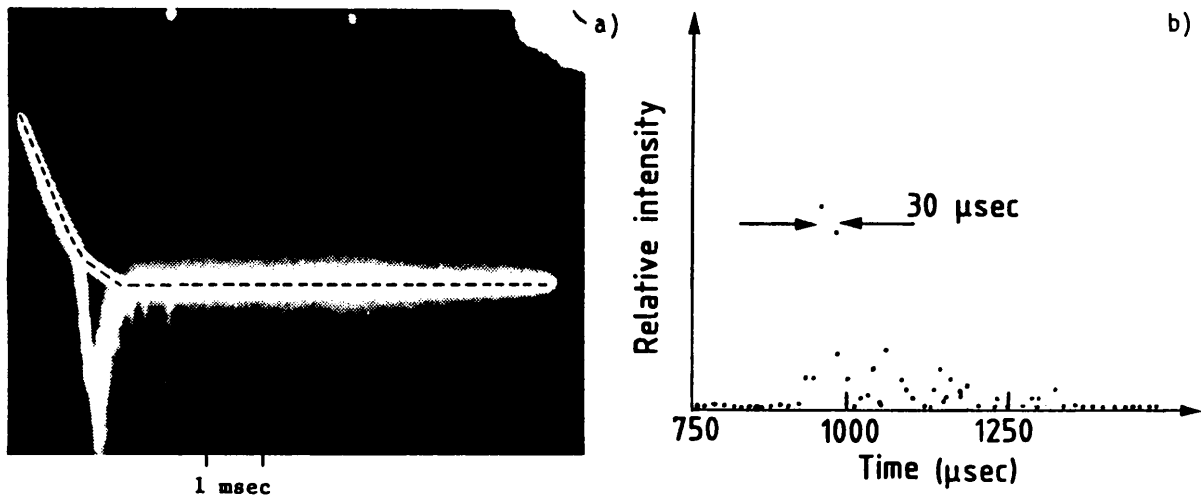polarity; b) $\rightleftarrows$ same polarity.

Fig. 7 a) The integrated light-pulse superimposed on the polarization pulse
(- - - -), showing the time correlation between light emission and crystal
polarization saturation. b) Time correlation between applied voltage to a
TGS crystal and the emitted light, using a PM as detector and a 4K
multichannel scaling system, of 10 µs bin width, at CERN-ISOLDE (Ravn).

The results obtained from such an investigation might well contain
relevant information, once they have been quantitatively understood. They
show very characteristic features such as: exponential slope and decay; the
UV light is emitted later than the visible light; the light intensity and
time structure depend on the environmental conditions, etc. According to
this and earlier investigations [9], this light emission is strongly
correlated to the crystal polarization (Fig. 7), so that it might well be
used as a trigger signal for the switching process.

The most surprising finding was to observe delayed light emission after
a 1.5 µs short aligning HV pulse to a TGS crystal (Fig. 8). Similar
observations have been made also with PZT as well as with PLZT. This is in
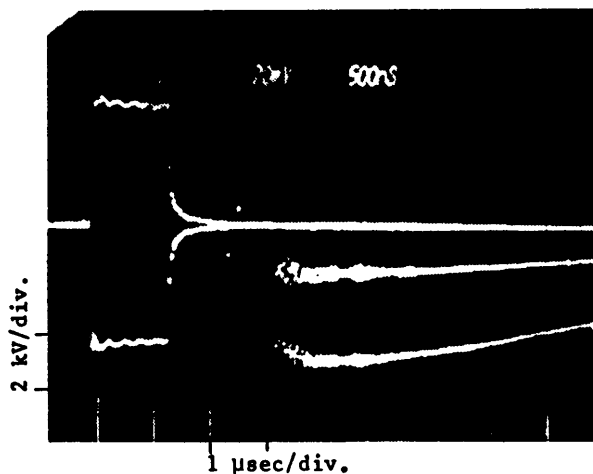

Fig. 8 Light emission (right) following 4 kV and 1.5 µs short HV pulses
(left) applied to a TGS crystal. The two light pulses correspond to the two
opposite polarization states.

full agreement with the picture of FE given in this work: the crystal takes the energy necessary for a complete or partial polarization reversal from the applied voltage, and it needs some time to finish the switching procedure, giving rise to all the phenomena associated with the switching mechanism. In other words, once the polarization reversal has started, the crystal has a kind of inertia continuing without any help from outside. This being so, it will have practical consequences.

7.2 Potential across the FE

We have measured the potential applied to a crystal (TGS) while using an HV pulsed generator. As shown in Fig. 9, the measured potential difference across the crystal is changing and decreasing at the beginning of the aligning pulse, which means there is a time-dependent dielectric
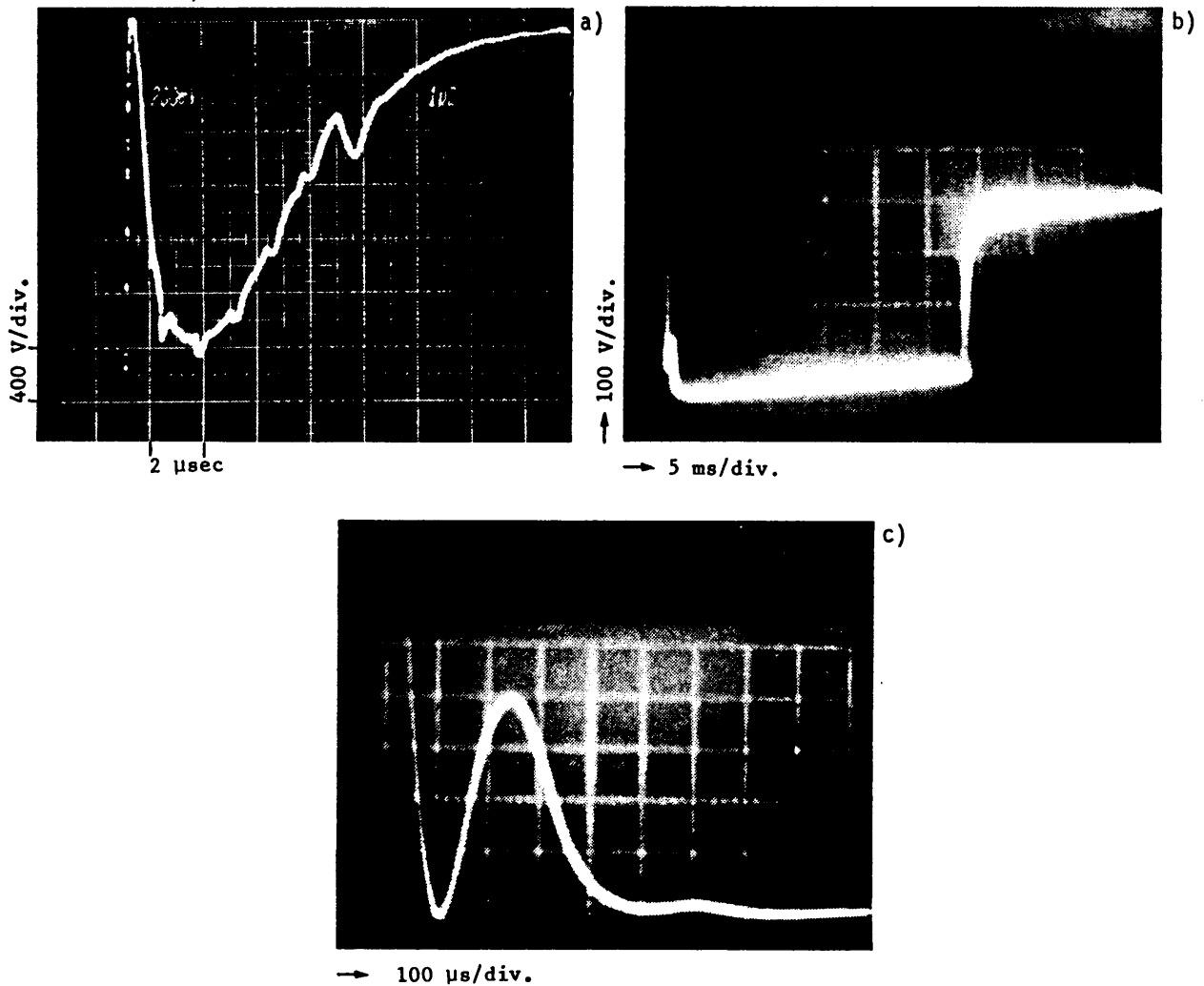

Fig. 9 Time-dependent potential across the FE, by applying a rectangular HV pulse: a) HV > 0; b) and c) HV < 0.

constant during polarization reversal, again in complete analogy to the FM. Therefore, this might be evidence for domains with the electric field inside being reversed during switching. The most important conclusion from this kind of measurement is that the time-dependent ε-curve is another, new experimental method for observing the switching process of an FE. The shape of this curve (rise- and decay-times, width, height), taking into consideration also the correlated emitted light (Fig. 6, 7), will give more insight into the dynamical behaviour of these materials. Investigations are under way, with a view to minimizing t_s for relatively large FE samples: $\approx$ cm^2 area and a few millimetre thickness. However, the t_s-values reached are rather encouraging.

CONCLUSION

We have already gained valuable and partly new information on the dynamical behaviour of FEs. Some problems appeared while working with such materials with the aim to establish very high electric fields. Care must be taken in order to avoid the compensation of the spontaneous polarization, so that such macroscopic fields can be obtained. The proposed solutions are promising and they are still an experimental challenge. A few 'simple' experimental approaches are now being carried out at CERN, although more sophisticated techniques may finally be necessary for a successful outcome. However, there is no theoretical objection to 100 GV/m electric field gradients, in particular on a microscopic scale. In a previous channelling experiment, very strong effects have been observed below the Curie temperature T_c. A steady contact with solid-state physicists[c] provides us with invaluable feedback. Results from previous measurements [4, 10] are surprisingly in full agreement with our most optimistic picture of FEs.

*   *   *

a) In collaboration with H. Gundel and H. Riege (CERN).

b) In collaboration with I. Giomataris (CERN), P. Petrof (LAL, Orsay), H.J. Besch, J. Séguinot and T. Ypsilantis (Collège de France, Paris).

c) P. Günter (Zürich), D. Siapkas (Thessaloniki), J. Albers and A. Klöpperpieper (Saarbrücken), L. Godefroy (Dijon), H. Schmid (Genève).

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