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**TIME-DEPENDENT ELECTRON EMISSION FROM FERROELECTRICS
BY EXTERNAL PULSED ELECTRIC FIELDS**

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ABSTRACT

Results of experiments investigating the influence of the amplitude, the growth rate, and the repetition rate of the exciting electric field pulses on the electron current and charge density emitted from ferroelectrics are reported. The behaviour of two types of lead-lanthanum-zirconium-titanate (PLZT) ceramics, 2/95/5 and 8/65/35, was studied. The temporal shift between the applied HV pulses and the emitted electron current pulses differed in both materials. Regular electron emission was observed at repetition rates of up to 2 MHz in the PLZT-2/95/5 material, showing that the recovery of the emitting sample back to the original state may happen in less than a microsecond.

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1. INTRODUCTION

During the last 30 years, thermal, electrical, optical, and mechanical procedures, or combinations of different methods have been tried out in order to bring about spontaneous polarization changes ΔP_s , and in this way to excite electron or light emission from ferroelectric materials. Most frequently, thermal procedures were applied. The electron emission excited by an external electric field was less often investigated [1-3]. Some studies of light emission induced by applying an alternating field or rectangular electric pulses are described in Refs. [4] to [9].

Several ferroelectric materials, among them barium titanate and chemically similar materials, were investigated. The emitted current density was, however, only 10^{-9} A/cm² and in most cases much less, independent of the applied method.

Recently, we developed an experimental method of fast polarization change, which is based on the application of a rapidly rising electric field pulse to thin disks of ferroelectric material via tightly fixed electrodes, one of which is grid shaped [10]. Using PLZT and PZT materials, with zirconium/titanate ratios of 95/5 and 65/35 and different lanthanum contents, we obtained emitted current and charge densities that are many orders of magnitude greater [10-12] than in the earlier cases mentioned above. These results were obtained in low-impedance circuits mostly at temperatures near the antiferroelectric (AFE)-ferroelectric (FE) and FE-PE (paraelectric) diffuse phase transition (DPT) regions (see, for instance, review papers [13-16] and Ref. [17]). The neighbouring AFE, FE, and PE phases coexist in a wide temperature region around the average AFE-FE and FE-PE phase-transition temperatures. A strong correlation between pyroelectric and thermostimulated current, on the one hand, and the emitted electron charge density, on the other hand, was found in these DPT regions [11, 12].

In the present paper, we have studied aspects of the influence on the electron emission of the electric field growth-rate of the applied external HV pulses, of the amplitude of the HV pulses, and of the repetition rate up to 2 MHz. The experimental results are presented and discussed.

2. EXPERIMENTAL TECHNIQUES

The ceramic PLZT-2/95/5 and 8/65/35 samples were prepared using a conventional mixed-oxide method [14]. The disk-shaped samples, of 1 mm thickness and 17 mm diameter, were polished and coated with evaporated gold electrodes ($\varnothing \approx 8$ mm), fully solid (SE) on one side and in the form of a striped grid electrode (GE) on the other side. The interconnected gold stripes, of 200 μ m width, were separated by bare stripes of equal width.

The samples were prepoled by applying a d.c. electric field of 2 kV/cm with negative polarity on GE. The sample was poled at 150°C for 30 minutes, and was afterwards cooled down to room temperature through the FE-AFE (sample 2/95/5) and PE-FE (sample 8/65/35) phase-transition temperatures ($\sim 142^\circ\text{C}$ and $\sim 130^\circ\text{C}$, respectively) under the field.

Figure 1 shows the top view (a) and side view (b) of the experimental set-up. The electrical circuit (Fig. 2a) consists of four capacitors charged to the desired voltage via a 1 M Ω charging resistor. Each capacitor is linked to a separate fast transistor switch (S_1 - S_4). The capacitors and switches are arranged symmetrically around a centre plate, on which the ferroelectric ceramic sample FE is mounted. When the switches are closed, a negative HV pulse is transmitted to the solid electrode (SE) of the FE sample. The pulse length is controlled via the transistor switches and was fixed at 150 ns. A resistor R_d damps the oscillations in the circuit. To reduce stray inductances, all connections were built in the form of strip-lines (SL). In order to vary the field growth rate, the switches could be triggered either individually or, simultaneously, together in any combination. The grid electrode GE was connected directly to earth. To avoid breakdown between SE and GE, the edges of the sample and the bare surface rings, outside the electrodes, were covered with an insulating layer. The electron-emitting-side GE was oriented towards a vacuum chamber ($\sim 10^{-6}$ mbar). The auxiliary

grid (AG) was grounded. This protects against noise from the HV pulse circuit. Also, the Faraday cup (FC), not shown in Fig. 1, was screened with a high-frequency protection grid. The FC is made of graphite. It was connected to the measuring circuits, shown in Fig. 2b, in order to measure the emitted electron current and charge.

The electric field growth-rate could be changed from 35 to 560 kV/(mm· μ s) given by the linear rise of the voltage waveform. The maximum current was limited to 120 A for each switch.

In the high-repetition-rate experiments, pulse bursts of up to 50 pulses of variable repetition frequency were applied. The maximum repetition rate of 2 MHz obtained with a double-pulse generator was limited by the performance of the switches and by the trigger pulse generator. The charge measured on FC was multiplied by a factor of 15 to take into account the transparency of the grid AG and the ratio of bare-to-electroded areas. The transparency of AG was calculated from charges measured with FC with and without AG. Because of a considerable pulse-to-pulse scatter in the current and charge amplitudes, which generally decreases when the applied voltage is increased, some of the experimental data given below represent an average over several tens of randomly chosen shots.

When measuring the emitted charge density as a function of the exciting electric field strength, we limited the field to a level below the breakdown value so as to avoid damage to the striped electrodes and to the emitting bare parts of the sample surface.

The bandwidth (400 MHz) of the available oscilloscope limits the measurement of the rise-time of the individual current pulses of which the whole emission burst is composed.

3. EXPERIMENTAL RESULTS

3.1. The electron emission from PLZT-2/95/5 ceramics

Figure 3a shows an example of the HV pulse waveform on SE together with the waveforms of the integrated emitted electron current measured on FC for a series of randomly selected shots. Figure 3b shows many short current spikes, which contribute to the whole emission process. There is a clear delay of about 40 ns between the moment at which the HV pulse reaches its full amplitude and the beginning of the emission process. This delay does not depend on the strength of the exciting electric field in the range of 20–40 kV/cm, nor does it depend on the field growth-rate within the limits of experimental error.

The influence of the exciting field growth-rate on the emitted charge density was studied by operating either one switch, or two, three, or four switches in parallel. In this way four different field growth-rates in the range from 75 to 560 kV/(mm· μ s) were achieved. The field growth-rate was measured in the linear rise of the pulse waveform of the HV traces (as shown in Figs. 3a,b). The small number of the growth-rate data that could be realized allowed the dependence of the charge density as a function of the field growth-rate (Fig. 4) to be established only approximately. After the initial increase of the emitted charge with growth-rate, a saturation-state seems to be reached.

The charge density emitted from PLZT-2/95/5 as a function of strength of the exciting electric field has been reported earlier [10, 11]. In this case, much longer HV pulses (1 μ s) with much smaller growth-rate were produced. Similar investigations were now repeated with HV pulses of 150 ns duration and field growth-rates of 400 to 560 kV/(mm· μ s).

The emitted charge density as a function of applied electric field strength is shown in Fig. 5. The electron emission starts at a threshold of about 20 kV/cm and increases, at first slowly and then rapidly, when the field strength passes 30 kV/cm.

The emission characteristics as a function of repetition rate of the HV pulses were first studied with a double-pulse excitation at 1 Hz, where the double-pulse spacing was between 500 ns and 10 μ s. Similar experiments were performed with a series of several tens (20–50) of pulses at frequencies up

to the 1 MHz range. Figure 6 shows a train of HV pulses with 1 MHz frequency and the waveforms of emitted current and charge pulses. The first emitted current pulse has generally a greater amplitude, and the next ones show scatter around an average value when the individual amplitudes of the whole HV pulse train are constant.

Both types of samples, PLZT-2/95/5 and 8/65/35, were prepoled prior to the electron emission tests. The layers close to the emitting surface contain an enriched concentration of donor-centres. These may act as sources of electrons, which are set free during P_s switching. This hypothesis was supported by the following experiment. The electrode GE was disconnected from ground after several field-excited emission acts, and an xy-recorder was connected to the SE and GE electrodes afterwards. A weak current, decreasing with time, was observed for several minutes after switching off the HV pulsing (Fig. 7). The current flow does not seem to depend on the number of previously applied HV pulses. The potential of the electrode GE was positive compared with SE. The donor-centres, which are emptied owing to the field-excited electron liberation, seem to be refilled by electron carriers arriving via bulk and/or surface conductivity. The refilling process of the centres located in the layers underneath the bare stripes of the FE sample takes place slowly, because of the rather small conductivity. A number of these layers near the striped electrode edges are filled more quickly by electrons injected directly from the electrode GE during or shortly after application of the HV pulse.

3.2 The electron emission from PLZT-8/65/35 ceramics

Figure 8 shows an example of the HV waveform across the electrodes of the FE sample, together with the charging current of the FE sample and the waveform of the emitted electron charge, a) with and b) without bandwidth limitation of the oscilloscope. In this and the following cases, the emitted charge was measured without the auxiliary grid (AG). Unlike the 2/95/5 sample (Fig. 3), there is no delay between the moment when the voltage amplitude reaches its maximum and the start of the electron emission. It seems that first the electroded part of the FE sample is charged like a capacitor (see the first part of charging current waveform). Then a virtual cathode is formed by released carriers (electrons), which are transported into thin surface layers close to the emitting bare surface areas of the FE sample. Most of these free electrons are emitted and new electrons are transported into the surface layers, probably by injection from the stripes of the GE electrode, while the charging current is still flowing. The current flowing through the bulk of the FE sample, the conductivity of which is increased, should also be considered. It has been shown in Ref. [18] that breakdown takes place, in the case of fully electroded FE samples, when fast-rising, strong HV pulses are applied.

When the field growth-rate was increased from about 35 to 200 kV/(mm · μs), the emitted charge also increased (Fig. 9). The dependence of Q_e on field growth-rate is similar to the behaviour of the 2/95/5 sample shown in Fig. 4.

The dependence of Q_e on the amplitude of the applied electric field pulses has already been reported for PLZT-8/65/35 ceramics in Ref. [12]. There, HV pulses of 350 ns duration and of a field growth-rate of about 10 kV/(mm · μs) were applied. The electron emission started at a threshold value of about 6 kV/cm. The same material, tested under the new conditions described above, shows a different dependence of the emitted charge density Q_e on the pulsed electric field strength E_p (Fig. 10). Curves 1 and 2 were measured when the amplitude of the HV pulses was gradually increased and then decreased. The much shorter (150 ns) and faster-rising HV pulses induce emission above a threshold of about 12 kV/cm. When the field is increased to 20–22 kV/cm, a minimum occurs in the Q_e(E_p) curve. For higher fields the emission increases sharply. The higher threshold field at faster-rising excitation current can be explained by a growing coercive field. Data quoted in the literature [18] show that the dynamic coercive field is considerably greater than the static coercive field. There are also big differences observed in the coercive field and in the remanent polarization

from individual grain to grain in the ceramic material [15]. This may explain the observation of a whole series of individual, separated current pulses of emitted electrons which are spread over a finite time interval linked to the growth time of the exciting field (Figs. 3 and 8b). Also, the large scatter of the emitted charge waveforms may be traced back to these effects.

For the PLZT-8/65/35 material, the experiment was performed also with HV pulsing at a high repetition rate. The charge emitted during the second pulse of the double pulse (spacing 1 μ s) was almost an order of magnitude weaker. Sometimes the second emission was lacking altogether or showed a strong scatter. Only for the larger pulse-spacing and higher excitation-field amplitudes was the electron current emission more reproducible.

4. DISCUSSION

The PLZT-2/95/5 material is characterized by a phase sequence AFE-FE-PE when the temperature is increased, whereas the PLZT-8/65/35 shows an FE-PE phase sequence. The coexistence of different phases in the diffuse phase transition regions AFE-FE and FE-PE, and the high donor-centre concentration, are important factors in the electron generation and emission processes. The experimental results obtained with these two materials are discussed separately because of their different phase sequences and their different material properties at room temperature, at which the experiments have been performed. There are, however, also some common features in both materials, which are relevant to the electron emission. Many PLZT materials, which are compositionally located near the boundary between the rhombohedral FE(R) and the tetragonal FE(T) ferroelectric phases or between the AFE and FE phases, exhibit, for instance, strong frequency relaxation characteristics of their dielectric response. In pure lead-zirconate (PbZrO_3), a fast-response dielectric relaxation was found in the microwave region [19]. The Zr-rich PZT and PLZT materials seem to behave in a similar way. Such materials are known as diffuse phase relaxers or relaxer-type ferroelectrics [14]. The FE(R) and FE(T) ferroelectric phases are referred to as 'soft' (low coercive field) and 'hard' (high coercive field) states, respectively. The FE(R) phase is electrically more easily switchable than the FE(T) phase.

The fluctuation of the composition in individual microregions and the differences in the internal electric fields and in the internal mechanical stresses, which prevail in their vicinity, lead to more or less diffuse FE-AFE/PE phase transitions, depending also on the Zr/Ti ratio and the La concentration [13-17]. The differences in the local phase-transition temperatures of individual microregions are especially large in the surface layers of the prepoled samples. The internal electric fields due to the space-charge polarization have a strong influence on the electrical switching behaviour in the FE domains located in the surface layers. The space-charge polarization field increases the number of the remaining metastable FE domains inside the AFE or the PE matrix especially in the surface layers. The FE state can also be induced in the originally non-ferroelectric matrices by applying sufficiently strong external electric fields. The specific microstructure and the phase composition allow small local regions to be switched successively when the external electric field is applied [14-16]. The shift of the AFE-FE phase-transition temperature towards lower temperatures under the influence of the external field is particularly strong in the case of PZT materials containing more than 3% mol Ti [20].

Many properties of the PLZT materials depend on the La concentration [14, 16]. Lanthanum leads to the creation of donor-centres in the bulk material. The La ions can replace Pb ions, and the charge balance in the ABO_3 structure is maintained by the creation of vacancies in the A or B sublattices. In Zr-rich materials, a majority of A-site vacancies predominates, whereas in Ti-rich materials mostly B-site vacancies are present [14, 16]. The high concentration of the donor-centres, especially near the emitting surface layer of the prepoled sample, can be considered as an additional source of electrons being liberated during ferroelectric domain switching.

Several authors assume that, independently of the type of electron emission excitation, the electrons are emitted from the negatively charged surface of the FE sample during a sufficiently rapid P_s change. The electrons are drawn from the donor energy levels, or even from the valence band, owing to excitation by a strong internal field that appears during the process of the rapid P_s change. The uncompensated charge, caused by the fast change of P_s and by the relatively long Maxwellian relaxation time of the space charge, gives rise to a strong electric field at the FE sample surface, which leads to the emission of electrons. For the restoration of the original sample state, the electron injection from the electrodes during the application of the HV pulse and after its end should be taken into account [5].

Some of the earlier experiments on electron emission excited by electric field pulses showed clearly the role of fast P_s changes in the surroundings of AFE-FE and FE-PE diffuse phase transitions [11, 12]. It was pointed out that different neighbouring phases, coexisting in the DPT regions, favour the electron liberation and emission processes. The FE domains located in the negatively charged surface layer are primarily responsible for the electron emission from the bare stripes of the sample surface.

The average AFE-FE and FE-PE phase-transition temperatures decrease and increase, respectively, under the influence of an external electric field [21]. This is also valid for the individual microregions with different phase-transition temperatures.

In the case of PLZT-2/95/5, the AFE state is thermodynamically stable at room temperature. This state transforms to the FE state when subjected to an external electric field of the order of 20 to 40 kV/cm [14]. The return to the AFE state is abrupt when the field disappears. The electron emission from the 2/95/5 material takes place just in this field range (Fig. 5).

The high-repetition-rate experiments indicate that FE domains induced by an electric field inside the AFE matrix may be responsible for the electron emission from PLZT-2/95/5, although the remanent metastable FE domains may also contribute. The fact that the two regions have different slopes in the $Q_s(E_p)$ dependence, shown in Fig. 5, is because they are based on these two different processes. The differences in the local AFE-FE phase-transition temperatures of individual microregions may be the reason for the different time-delays of the individually emitted short current pulses within one emission pulse (Fig. 3). The measured minimum delay of 40 ns could be the time that is necessary for the formation of FE nuclei and the domain-growth by domain-wall motion. The electric field that results from the external and internal fields is enhanced in the surface layers of prepoled samples. Consequently, the electrically induced FE domains appear first of all within this layer. Removal of the external electric field allows the material to relax rapidly and return to its non-polar state. This may be an explanation of the fast recovery of the PLZT-2/95/5 in the high-repetition-rate experiments.

In the case of the PLZT-8/65/35 material, the FE phase with rhombohedral symmetry is thermodynamically stable at room temperature [14]. Therefore, the material behaves in the 'soft' (low coercive field) state. The investigation of pyroelectric Barkhausen pulses originating from individual domains in the rhombohedral phase of $PbZrO_3$ single crystals proved the existence of twin domains with compensated antiparallel P_s vectors [22]. Spontaneous polarization reversal in one part of the twin domains occurs via the sideways motion of 180° domain walls, which are far more mobile than other types of domain walls [18].

In the prepoled sample, the pairs of the antiparallel P_s vectors are — at least partially — oriented perpendicularly to the emitting FE surface. When one of these P_s vectors starts to reverse under an externally applied HV pulse, the resultant spontaneous polarization appears almost instantaneously in the twin domain. The negligible time delay between the HV pulse rise and the electron emission may be a consequence of the fast reversal of one half of the twin domain. When the reversal process in the twin domains is over and emission has taken place, the single domains are partially fixed by the

internal electric field in the thin surface layer and by screening processes. The emission process cannot, therefore, be repeated so quickly with high repetition excitation, unlike in the case of the 2/95/5 material. However, the twin domains are again formed under the influence of the internal field raised by the remanent spontaneous polarization in part of the single domains. If the HV pulse repetition rate is in the range of 1 Hz, the electron emission takes place regularly at each pulse, but the scatter in total emitted charge amplitude is much bigger than with the PLZT-2/95/5 material.

5. CONCLUSIONS

The electric-field-excited electron emission from ferroelectric materials is a new tool for investigating the rapid switching (reversing) of the spontaneous polarization and also the dynamics of ferroelectric domains. The method can be utilized in fundamental investigations of both existing ferroelectric domains and those that are created by external electric fields in the antiferroelectric or paraelectric matrices.

The electric-field-excited emission of electrons from the surface of ferroelectric materials has been studied as a function of growth-rate of the exciting field, at high repetition rate. The results of the measurement show that very fast processes inside the materials with coexisting ferroelectric, paraelectric, and antiferroelectric phases are responsible for the rapid ejection of the electrons from the surface. The different temporal behaviour of the two materials that have been investigated in the experiments might be related to different types of relaxers existing in these materials. The absolute speeds of the P_r change, which have been confirmed experimentally, do not seem to be the final natural limits, but are the values determined partially by the experimental set-up and by the methods applied.

The measured data are nevertheless of great importance for evaluating the field-excited electron emission from ferroelectrics for practical applications. The dependence of emitted current amplitude on field growth-rate should be linear in an electrical circuit as used in these experiments, provided the stray inductances are zero. A finite inductance in series with the FE sample will necessarily lead to a saturation of the emitted current, when increasing the storage capacity. This effect has been observed. For reaching even faster switching times, especially in the subnanosecond range, extremely low-impedance, low-inductance circuits must be designed. Strongly powered electronic switching arrays will be required for performing fast spontaneous polarization changes in large ferroelectric emitter arrangements for intense electron beam sources applied to power switching and preionization, or in the field of accelerators.

The high recovery rate of certain ferroelectric materials after an emission pulse allows for the design of very rapidly pulsed systems in the megahertz range applicable in microelectronic and medium-size power-switching systems. Attractive features are the 'self-resetting' of the emitter after each emission and the fast restoration of charge to the emitting surface.

One also may conclude that the emitted electron beam consists of a large number of individual microbeams, each coming from a different domain. The homogeneity will hence be improved when shorter pulse times are reached. The best results could be obtained with single-crystal emitters produced by thin layer techniques. This could be more easily reached on a microelectronic scale.

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Figure captions

- Fig. 1: The experimental set-up: a) top view, b) side view.
- Fig. 2: The electrical circuits: a) high-voltage pulse excitation circuit, b) measuring circuits.
- Fig. 3: Typical oscilloscope traces of voltage (bottom of Fig. 3a) and emitted charge waveforms (top of Fig. 3a) and emitted current waveform (top of Fig. 3b) for the PLZT-2/95/5 sample excited by a HV pulse (bottom of Fig. 3b).
- Fig. 4 The dependence of the emitted charge density as a function of the field growth-rate for three selected strengths of the applied electric field for the PLZT-2/95/5 ceramic.
- Fig. 5 The emitted charge density Q_e as a function of the applied electric field strength E_p .
- Fig. 6 The typical waveforms of the voltage (bottom) and a) emitted current or b) charge (top) when a burst of 1 MHz pulses are applied to the PLZT-2/95/5 sample.
- Fig. 7 The current $I(t)$ flowing between the SE and GE electrodes after a HV pulse has been applied and the electrodes have been directly connected.
- Fig. 8 Typical waveforms of the voltage (bottom), of the charge, and of the current charging the FE sample (middle) with (a) and without (b) bandwidth limitation for PLZT-8/65/35 ceramic.
- Fig. 9 The emitted charge density as a function of the field growth-rate for three selected strengths of the applied electric field, for the PLZT-8/65/35 ceramic.
- Fig. 10 The emitted charge density Q_e versus the electric field strength E_p , for PLZT-8/65/35. This dependence was obtained when the amplitude of the HV pulses was first increased (1) and then decreased (2).

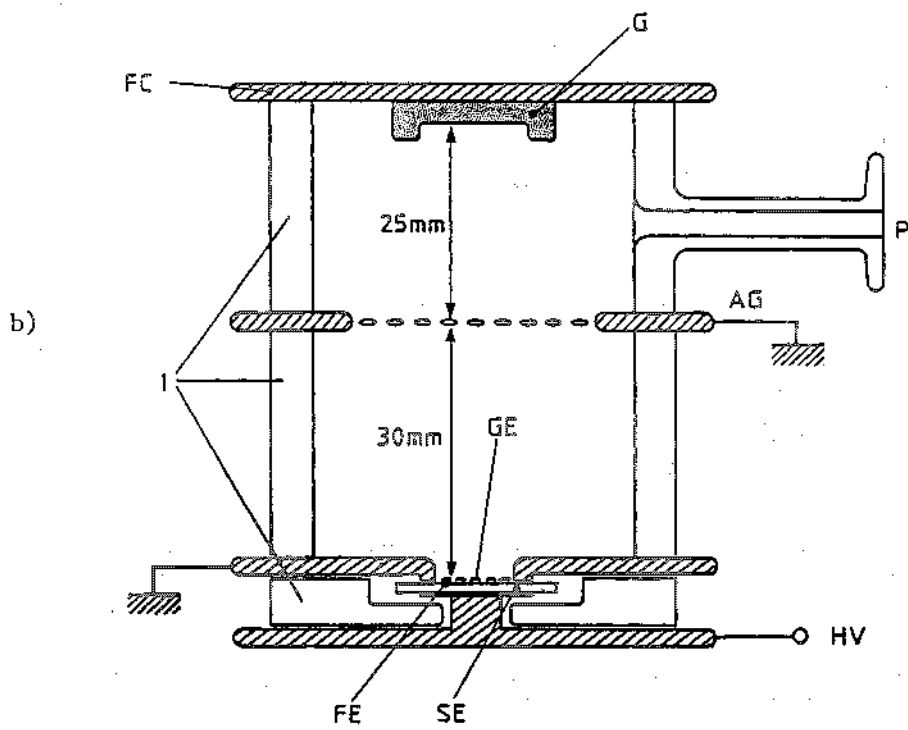
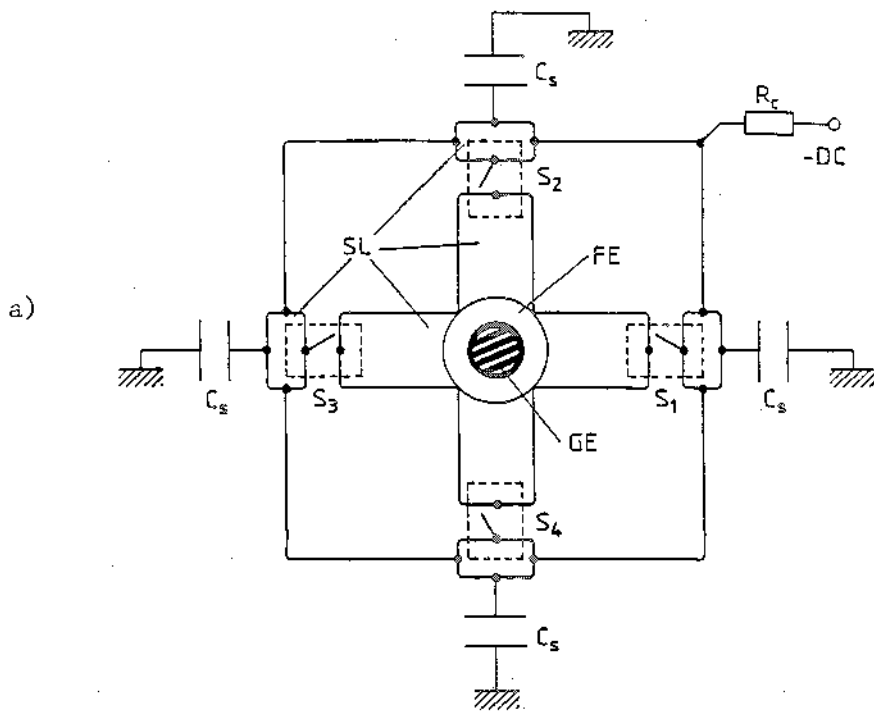


Figure 1

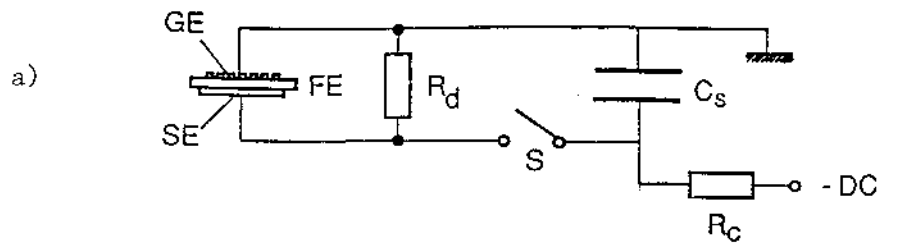
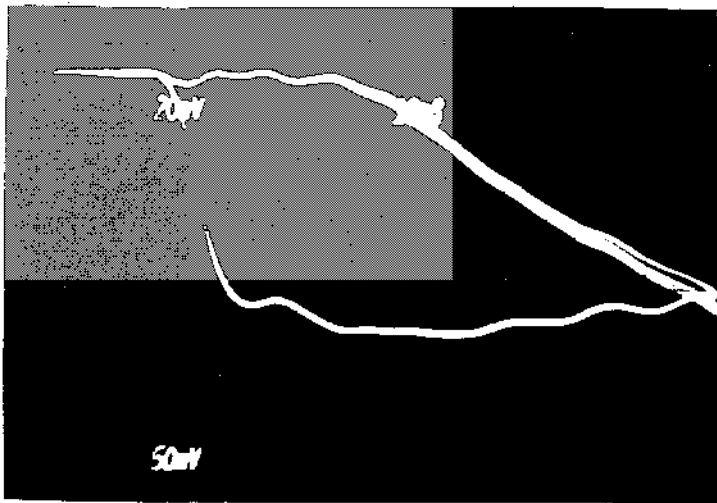
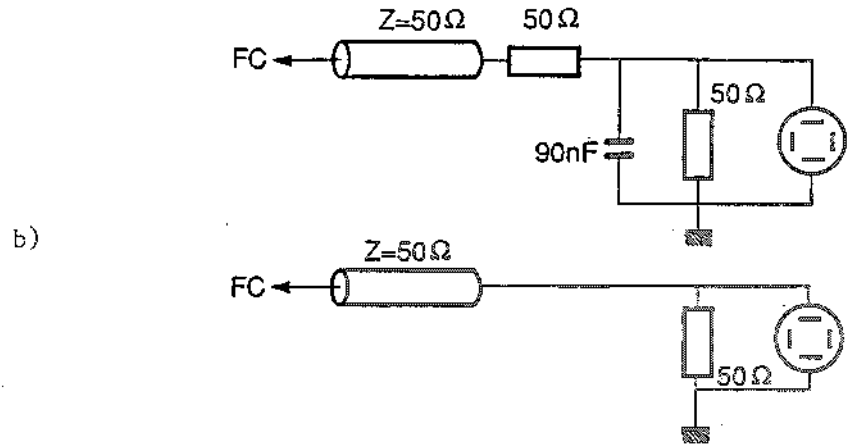


Figure 2



a)

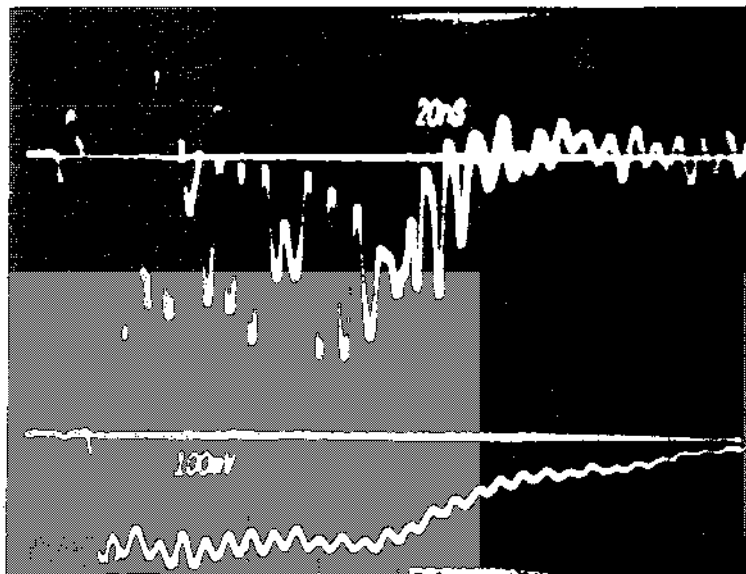


Figure 3

b)

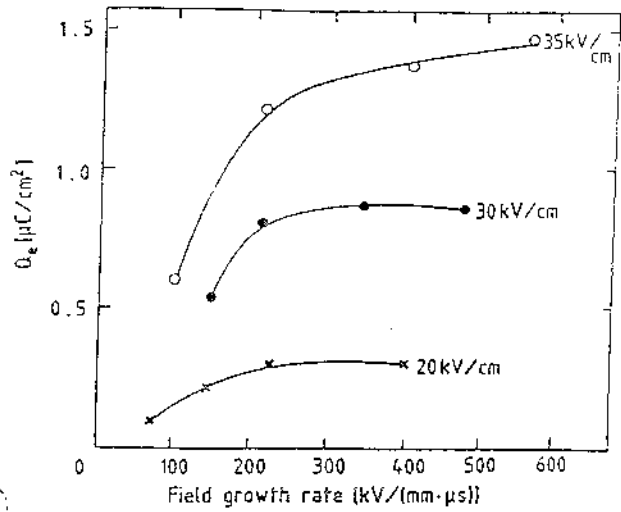


Figure 4

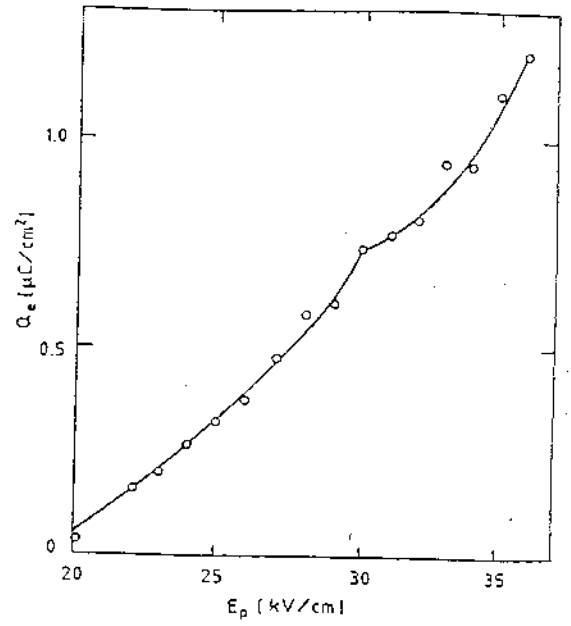
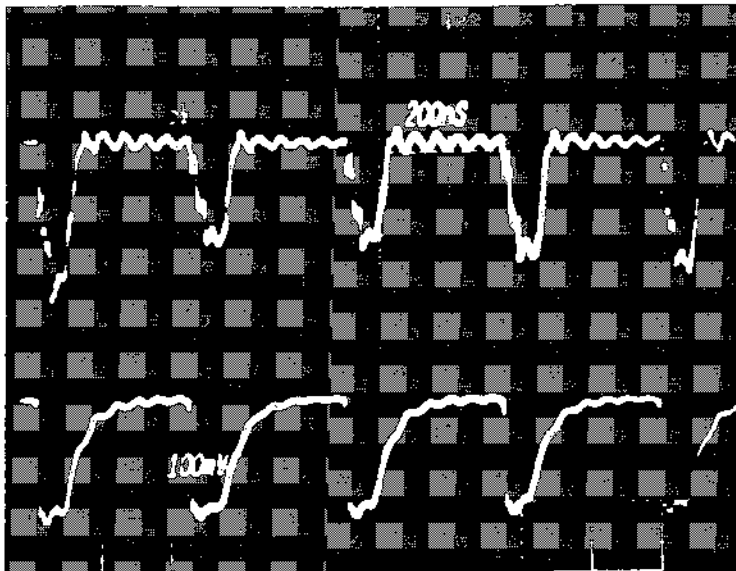
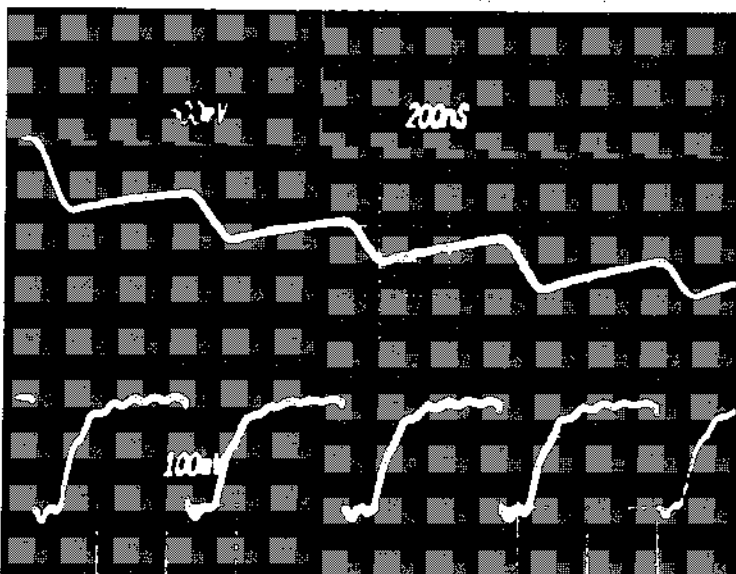


Figure 5



a)



b)

Figure 6

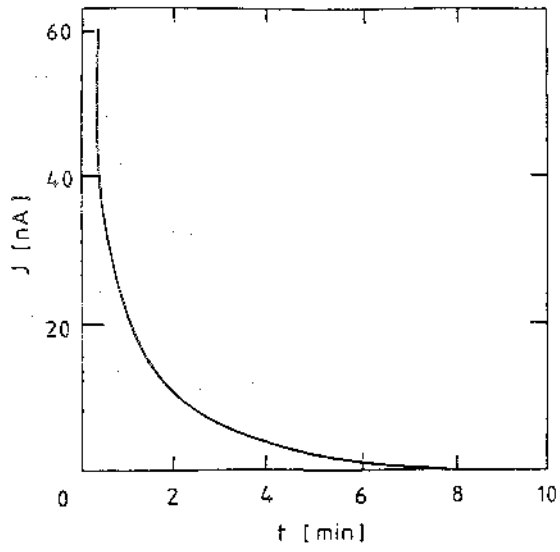
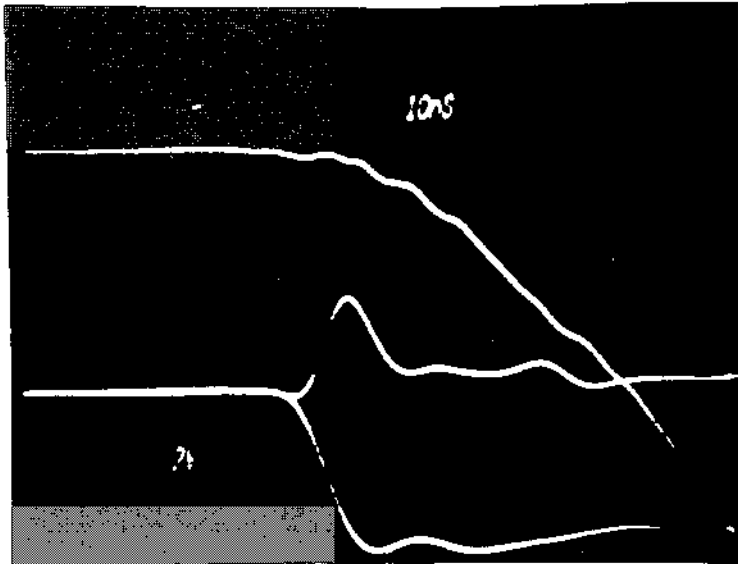
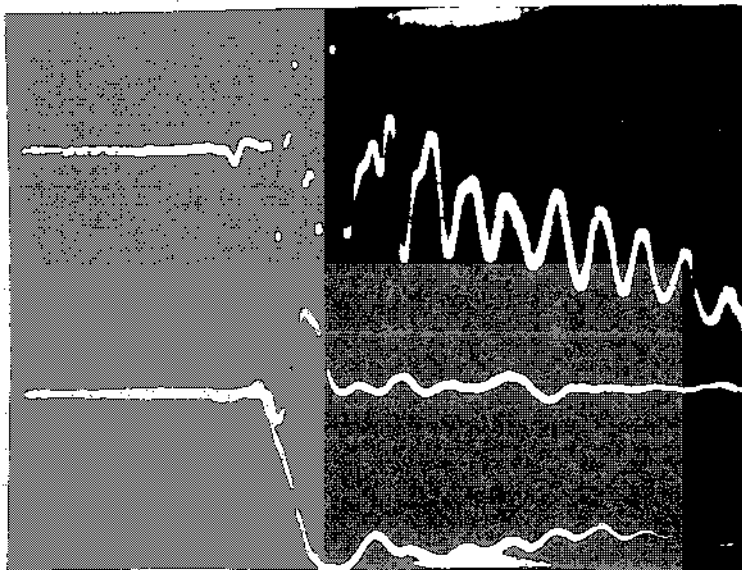


Figure 7



a)



b)

Figure 8

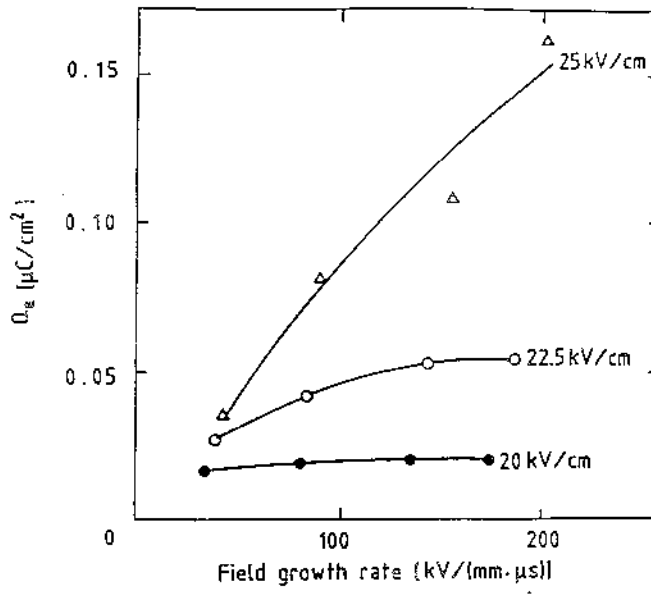


Figure 9

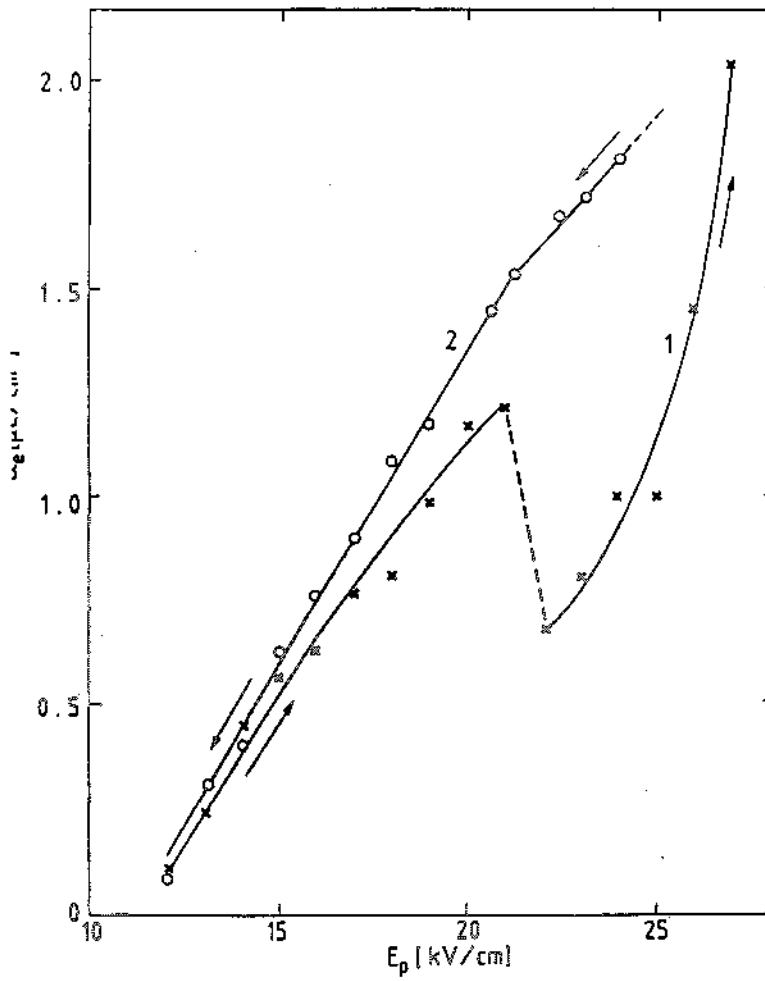


Figure 10