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**Production of Intense Electron Beams with Ferroelectric Materials**

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**Abstract**

Ferroelectric (FE) emission is based on the macroscopic separation of electric charges and on the self-emission of electrons under the influence of their own space-charge field. Surface charge densities of  $\mu\text{C}/\text{cm}^2$  can be generated by submicrosecond changes (switching) of the spontaneous ferroelectric polarization  $P_s$ . Polarization switching can be induced by fast electrical-field or mechanical-pressure pulses, and by thermal heating. Also irradiation with short laser pulses at wavelengths ranging from uv to ir leads to self-emission of energetic electron beams. The most attractive features of FE cathodes are high emission efficiency, robustness and easy manipulation.

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## 1. Introduction

Ferroelectrics are ideal materials for inducing the macroscopic separation of high charge densities and, consequently, for generating high electric fields, which accelerate any charged particle in the neighborhood including the FE-generated charges themselves ('self-emission'). Electron emission from FE material was noted in 1960 by Miller and Savage [1]. The experimental observation of weak electron emission as a consequence of electrically induced, spontaneous polarization reversal in FE materials was first reported by Rosenman et al. [2] in 1984. In 1988 Gundel, Handerek, Riege and Zioutas were able for the first time to produce "strong" electron emission from FE materials with current densities of more than  $100 \text{ A/cm}^2$  in the absence of any external extraction field. This strong electron emission can be excited, for example, by submicrosecond polarization switching with high-voltage pulses [3–8], applied via semi-transparent grid electrodes to a disk of FE material. The high, free-surface-charge densities, which provoke the pulsed 'self-emission' of intense electron beams, are mainly generated by spontaneous polarization switching of domains in the bulk and at the surface of the FE material. Hence, the ferroelectric emission very much resembles the spark emission from dielectric surfaces induced by friction and subsequent charge separation. It must however, be distinguished from the electron emission produced by discharges caused by the strong air-gap fields between an electrode and an adjacent insulator surface element, called metal-insulator emission, which only works with an extraction potential from the resulting surface plasma [9–10]. Also laser-induced electron emission from FE materials [11–12] is based on spontaneous polarization switching, and therefore differs strongly from the classical photoemission. Meanwhile, R&D on FE emission phenomena and related applications has started in several countries [13–20]. The purpose of this paper is to describe the actual status of the R&D on electron emission from FE and of some applications envisaged.

## 2. Principles of FE Emission

The conventional methods of electron emission (for example, thermionic, secondary, field and photoemission) require high extraction potentials in order to separate the charges from the surface of the emitting medium. In FE materials macroscopic (mm or more) separation of charges can be induced by several methods. Figure 1 gives a simplified idea of the emission process from an FE cathode. The FE layer is represented by a single-domain structure with two possible directions of the spontaneous polarization vector  $P_s$ , [8]. The originally compensated, spontaneous polarization is rapidly changed. This fast, submicrosecond polarization switching is the trick that is required to generate the macroscopic charge separation on the two opposite surfaces of an FE sample. The resulting unscreened space-charge fields are so high that self-emission of electrons takes place from the negatively charged free surface areas [8], [11]. In FE single crystals a multi-domain structure is present with several possible orientations of  $P_s$ , with respect to the FE sample axis and to the direction of the applied switching field. An even higher multiplicity is found in FE ceramics which consist of many differently oriented grains, subdivided into many smaller domains. For each domain a hysteresis curve is valid as shown in Fig. 2. For multi-grain and multi-domain materials an average hysteresis curve can be defined and measured, but high surface charge densities can be created from any domain at either surface, when the necessary electric field conditions are reached. Generally the first domains, which are switched and which start to emit, are those near to the edges of the grid electrode. The switching of an isolated domain may induce switching of further domains in the neighbourhood. The FE phase of most ferroelectrics is existent only in a limited temperature range. Especially in the PZT and PLZT ceramics used in most of the R&D on FE emission, the phase-transition takes place in a wide temperature range (diffuse phase transition). In other words, in many of these materials FE and non-FE phases may coexist at a given temperature, which, under the influence of electric fields or mechanical stress (see Fig. 3 a,b), can quickly undergo a transformation from a non-FE to an FE state or vice versa.

The result of such a fast phase transition is the same as that caused by a rapid  $P_s$  reversal. Both modes of polarization change may actually happen in such PLZT ceramics simultaneously and induce emission. The average spontaneous polarization of a virgin FE sample is zero. By applying an electric d.c. field at temperatures above the phase transition temperature the FE cathode can be prepoled: e.g. a resultant average polarization can be created perpendicular to the surface through the combined action of an internal space charge field produced during the prepoling and by the externally applied d.c. or a.c. field. Prepoling leads to a shift of the "averaged" hysteresis curve against the former, symmetric position with respect to the origin [21] and allows to switch FE cathodes repetitively without applying resetting pulses, since the altered polarization is drawn back by the internal prepoling field to its original state. By prepoling with a minimal electric dc field PLZT ceramics, which by composition may be non-FE at room temperature, a finite amount of FE phase can be generated. A threshold field of the order of the coercive field strength  $E_c$  has to be applied before emission starts. The field rise-time up to the threshold amplitude for  $P_s$  switching must be very short (submicrosecond), otherwise the charges produced by the switching can flow away over the surface or through the bulk of the FE material. The threshold fields measured with nanosecond excitation may exceed the coercive fields under dc conditions by two orders of magnitude (1 to 100 kV/cm). In order to achieve fast switching of  $P_s$ , it is not necessary to completely reverse  $P_s$  as indicated in Fig. 1, but to change only a part  $\Delta P_s$  of  $P_s$  either by moving under mechanical or electrical stress along the hysteresis curve (Fig. 2), by creating or annihilating FE domains with temperature changes, or by crossing phase boundaries when applying electric-field or mechanical-pressure pulses (Fig. 3, [3]). The mechanically induced self-emission by fast phase transitions from FE had been applied longtime ago for the spark generation in fire-lighters and demonstrates clearly the differences of emission from dielectrics and from FE. Negative surface charges are always formed on the surface where the positive polarization charge is decreased. This is in contrast to what happens in metal-insulator emission from FE, which functions only, when the surface electrode is negatively charged, hence, when a growing positive polarization charge is formed on the surface of the FE sample, [9–10] which reacts then like a dielectric with high dielectric constant. Gridded dielectric (e.g. glass) cathodes of the same construction as the FE cathodes described in reference [5–8] may induce electron emission after the formation of a local arc plasma by the air gap fields between the metal edges of the metal grid and the dielectric surface. Strong metal-dielectric emission can, however, never be observed without an extraction potential. Contrary to FE cathodes metal-dielectric emitters are not capable to produce self-emission. This self-emission is, hence, the main feature of the true FE emission and may be used as a definition of the phenomenon. Already at relatively modest average FE emitted electron current densities also on gridded FE cathode surfaces plasma formation may occur and plasma-assisted emission is interfering with FE emission in a positive or in a negative manner.

### 3. FE Emission by HV Pulses

FE electron emission (FEEE) by fast polarization switching when applying HV pulses to disk-shaped FE samples via gridded electrodes, has been used in most of the experiments that have been carried out since the discovery of strong FE emission. Figure 4 shows the emitted current and charge waveforms observed in vacuum FE emission induced by a negative HV pulse to the rear electrode of an FE sample previously prepoled with a negative d.c. voltage applied to the grid electrode [8]. The collector electrode in front of the FE, which captures the emitted current is at the same potential as the emitting FE grid electrode, hence, there is no extraction field. However, the emission is working also in different configurations. Strongly emitting FE cathodes may be even positively pulsed on the emitting grid electrode: then the low energy electrons are retained at the surface, whereas electrons with higher energy than the applied switching potential can escape and can be measured. Contrary to classical emission methods the processes of electron generation and acceleration can be separated in FE emission. When optimized FEEE with minimum mechanical and electrical fatigue is aimed at,

the main technological problems to be mastered, are the choice of composition, the production and the preparation of the material, the processes of reliably electroding and insulating the FE cathode surfaces, and the proper choice of the prepoling procedure. The HIV pulses needed for the  $P_s$  switching of PLZT samples correspond to field strengths above 1 MV/m. The HV pulse length should be between 0.1 to 1  $\mu$ s. The HIV switching voltage amplitude may be decreased according to FE sample thickness. The lowest pulse voltage reached so far by  $P_s$  switching of 30 to 45  $\mu$ m thick PZT samples was 75 V [16]. Even lower excitation fields seem possible with thinner layers; however, a limit will be reached when the polarization fields formed after switching no longer exceed the potential barrier fields at the FE surface. Since typical FE emitters have capacities of 0.1 to 100 nF/cm<sup>2</sup> depending on thickness and material, high-current switching pulse amplitudes of the order of  $10^2 - 10^3$  A are needed per cm<sup>2</sup> when beams with large cross-sections have to be generated. The importance of fast rise-times of the exciting HV pulses has been reported in [6]. Emitted current densities with amplitudes above 100 A/cm<sup>2</sup> and pulse lengths usually shorter than the excitation pulse length, but strongly dependent on the FE material type, have been measured without any extraction potential. The emission is independent of the quality of the vacuum, but can be enhanced in the presence of a low-pressure gas or of a plasma [23]. In practice no upper limit of emitted current density is observed. The reason is the formation and expansion of a plasma layer across the emitting surface when the local current density between the grid electrode and the nearest reversed FE domains exceeds a threshold, which, in the case of FEEF, can be far below 100 A/cm<sup>2</sup> average current density. The plasma formation is visible on the grid electrode during switching as a homogeneous glow or even as a large number of small, concentrated sparks. Strong plasma-assisted emission can have destructive consequences, especially in the presence of a high d.c. extraction potential. Even in vacuum a total electrical breakdown can be ignited between the FE and the collector electrode. But a weak surface plasma may have several beneficial effects, such as neutralizing the space-charge (favourable for beam emittance), forming a reservoir for fast recharging of the unscreened FE surface after switching, and enabling the homogenization of the emitted electron beam. A maximum repetition frequency of 2 MHz of FEEF was achieved with a prepoled PLZT 2/95/5 sample [6]. The repetitive pulsing worked without the application of a reset pulses. Rapid recharging via the aforementioned surface plasma, and fast reversal on the hysteresis loop returned the sample back to its original state.

#### 4. Laser-Induced FE Emission (LIEE)

As in classical photoemission the waveform of the electron beam current emitted from an FE photocathode (pc) and the light-intensity pulse have the same shape down to the picosecond pulse length range. LIEE was first believed to be a special case of conventional photoemission [24–25]. But this model had to be abandoned when strong self-emission of energetic electrons by laser irradiation of FE cathodes was observed in absence of any extraction field [8], [11–12] and [26]. Kinetic energies of the emitted electrons in excess of 10 keV have been measured. The observation of kinetic energies of this order of magnitude can be only explained by a macroscopic charge separation in the FE sample induced by polarization switching through the laser pulse. LIEE therefore resembles FEEF and differs totally from classical photoemission. The laser-induced self-emission appears only after a previous  $P_s$  switching for a limited number of laser pulses and can be regularly re-established with interleaved HV switching. The strength of "normal" LIEE (with d.c. extraction field) can be influenced by several factors, such as by the prepoling of the FE pc, by previous polarization switching with HIV pulses, or by interleaved laser irradiation without extraction voltage. Enhancement of normal LIEE up to a factor of 200 was observed after previous polarization switching with HV pulses [25]. The enhancement factors decrease with rising extraction field. Enhancement and self-emission require both a regular, interleaved polarization switching in order to be maintained at a constant emission level. Without such a refreshment of the FE polarization and surface charge density state the enhanced emission decays after some tens or hundreds of laser pulses to the normal LIEE emission level. In the same way as for

FEEE and contrary to classical photoemission, a threshold laser energy is observed in laser-induced self-emission, below which emission stops (Fig. 5). The term of quantum efficiency (QE) can be only used artificially in LIEE for comparing FE with standard pc's. Physically, however, it is meaningless, since LIEE is not determined primarily by potential barriers and should as such, function with incident electromagnetic radiation of any wavelength, provided a significant part of incident energy is absorbed inside the FE pc. We define the efficiency for LIEE as (emitted charge/nC)/(laser beam energy/mJ). Down to a timescale of less than 0.2 nanosecond no time-lag could be observed between laser and electron-beam pulse. Self-emission experiments by excitation of FE pc's with 140 fs laser pulses [26] confirmed that polarization switching of FE material can be achieved in less than 1 ps. In normal LIEE experiments with different laser pulse lengths it was shown, that the emission efficiency is increasing with laser power density by several orders of magnitude. This holds especially for longer wavelengths in the visible and the infrared range. Figure 6 (from [26]) shows the emission efficiency at 30 kV extraction voltage of one and the same PZT 95/5 FE pc doped with 2%  $Cr_2O_3$ , as a function of laser power density obtained with 5 ns, 40 ps and 140 fs long laser pulses at 266, 355, 390, 532 and 780 nm wavelength and with laser spot diameters on the FE target ranging from 0.5 to 5 mm. The decay of efficiency, which is observed at higher laser power density levels, and which happens at a lower power density level for shorter wavelengths, has been traced back to the influence of space charge at the given extraction voltage of 30 kV applied in the experiments shown in Fig. 6.

## 5. Applications of FE Emission

The main advantages of FE cathodes in technical applications [30] are, apart from high emissivity, the easy handling and the reliability during operation. For most applications considered up to now the specified charges and currents could easily be produced with FE emission. More important is the robustness and longterm stability of FE cathodes made from ceramic PLZT material, which compares well with that of metallic cathodes, while producing much stronger emission with FEEE, as well as with LIEE. FE cathodes can be manually touched without any special precautions and without the danger of degrading the emission efficiency. FE cathodes can be transported through air without problems of contamination, and they do not need good vacuum for operation. FE cathodes can be operated in an environment of low-pressure gas or plasma. Especially for high-power, cw applications, the features of a good conversion efficiency of primary wall plug energy into electron beam energy, as well as the high-repetition-rate performance capability of ceramic FE materials are interesting requisites. In high current density applications the macroscopic charge separation and the strong polarization fields favour the beam transport away from the emitter surface. Electron generation and acceleration may be separated. The electrons are ejected from the FE surface with an initial energy of the order of the applied switching potential, so that the Langmuir-Child limitation can be exceeded by several orders of magnitude [14] and magnetic focusing of the electron beam is feasible, where it leaves the FE surface. For the production of short electron beam pulses with FE pc's laser systems featuring the shortest possible pulse lengths, e.g. in the femtosecond range, are most suitable. Emission efficiencies may be obtained, which compare with those of semi-conductor cathodes at long laser pulse lengths, but which offer a much more stable and reliable operation of FE pc's under high average power conditions. Laser pulses of 140 fs length and with a power density up to  $2 \text{ TW/cm}^2$  have been repetitively (1 kHz) applied to a doped PZT pc [26] without causing any visible damage on the FE surface. The same pulses when directed onto the metallic support of the FE pc, burned spots into the metal surface and provoked vacuum breakdowns. The efficiency increase with power density measured on FE pc's, is especially marked at longer wavelengths in the visible and infrared region. This enhancement of FE emission allows to circumvent the use of uv laser light, which is required for metallic or CsI-type cathodes, when dense and short electron bunches have to be generated. Amongst the factors, which allow to control the FEEE and LIEE efficiency of FE cathodes, the composition and the preparation (pre-poling) of the cathode material may have strong influence on the emission. A reliable operation is only

guaranteed with ceramics of lowest porosity and with small piezoelectric coefficients, which render mechanical fatigue less effective. In LIFE applications the absorption edge of the FE material should be raised towards longer wavelengths, e.g. by material composition and or by doping [31].

### *5.1 Accelerator applications*

Already in the early stages of development, FEEE-excited FE cathodes were envisaged as a replacement for thermionic cathodes in electron guns for the injectors of accelerators. Higher reliability, robustness and emitted current densities, more flexibility in spatial and temporal beam pulse shaping, and easier transportability are the main technical advantages. Another example for dedicated FE electron sources are the electron-beam triggers of the high-power, low-pressure pseudospark switches of the pulse generators for the future LIIC beam dumping system at CERN [32–35]. Figure 7 shows a two-gap version of the switch with one FE trigger for each gap. The FE cathodes emit annular beams of 10 - 100 A (60 A/cm<sup>2</sup>), which are amplified by applying an extraction potential of 300 V and a low-pressure deuterium atmosphere. Both beams initiate, with nanosecond precision through each gap a 60 kA discharge from a storage capacitor charged to a voltage up to 35 kV [34]. The switching and emission efficiency and the long-term reliability of the FE cathode under the very difficult environmental conditions of contact with a hot plasma and metal vapor atmosphere, have proved to meet the life-time specifications in many prototype tests. FE emitters may be further used as short-pulse, laser-driven FE pc's. At wavelengths of up to 355 nm charges of a few nC per pulse have been reliably and reproducibly generated during several 10<sup>6</sup> shots without any degradation of the cathode surface as reported in [8] and [36–37]. Very promising is the development of fs-laser-driven FE pc's at wavelengths in the visible or infrared range. Experimentally it has been shown that the electron beams emitted from FE by FEEE can induce diffuse vacuum discharges and sputter ions from a target to form a strong ion-beam source [38]. This procedure is very efficient, when some kind of ion-beam neutralization is applied. In reference [39] enhancement of a vacuum-arc ion source is proposed, which is achieved by further ionizing and neutralizing the ion beam with a counter-moving, FEEE-generated electron beam. The increase by orders of magnitude of the output current of a laser-ion source has also been shown experimentally with an FE generated electron beam sent from the outside into the source [40]. By inductive acceleration the ion beam pulse can be accelerated in the presence of the neutralizing electron beam. FE-beam induced beam neutralization could find an application in future high-current ion linacs, for example in inertial confinement (ICF) particle beam fusion drivers, to transport the required beam currents in a small number of accelerator beam pipes and to succeed in beam collision at the fusion target without space-charge blow-up.

### *5.2 Industrial applications*

Though the FE emission development work at CERN is fully directed towards accelerator applications, FE electron emission can be and is already applied in a growing number of other technical fields. It seems attractive to use FE cathodes in high-power GHz microwave tubes, such as x-band klystrons or gyratrons, which could profit from the higher reliability and stronger emitted current densities of FE cathodes compared to tubes with thermionic emitters. High-power, quasi-d.c. electron beams produced with interleaved FE cathode systems running at high repetition rate could represent an interesting alternative to thermionically driven electron guns in machining devices or in electron beam driven chemical reaction chambers, e.g. for exhaust gas transmutation. The largest number of applications can be imagined in electronics, microelectronics and vacuum-microelectronics. Thin-layer FE cathode arrangements could replace the widely developed micro-tip field emitter arrays (FEA) [41–43]. Microelectron guns [44] producing microscopic electron beams by FEEE and/or combinations with multi-microlaser arrangements (LIFE) and nanotechniques may lead to revolutionary new devices. Though the micro-tip technology has made striking advances in fabrication and performance, FE cathodes with a thickness of more than 10 μm can reliably deliver one order of magnitude higher current densities combined with the known advantages of robustness and modest vacuum require-

ments, especially in high average power environments. Industrial work [16] has started on the development of ultraflat TV screens using FEE from thin FE layers (Fig. 8). A similar technique may be applied to the generation of high average GHz power from large-area thin FE layers as it was proposed earlier with laser-driven, ribbon-like pc's (lasertron) or with large FEA's [45-46].

## 6. Conclusions

The methods of generating intense electron beams with FE cathodes are attracting increasing attention. The simple procedures by which FE cathodes can be produced, the easy way such cathodes can be handled and the robustness under severe operating conditions have favoured a fast technology transfer from the stage of first proof-of-principle experiments to technically useful applications in the fields of accelerator techniques, pulsed power technology, electronics and vacuum-microelectronics. This seems to be the beginning of a wider expansion into various fields of electron-beam devices. In many applications where high average power electron beams are involved, FE cathodes could replace thermionic cathodes, normal field emission cathodes and FEA's, thanks to the capabilities of high current density emission and long operational performance.

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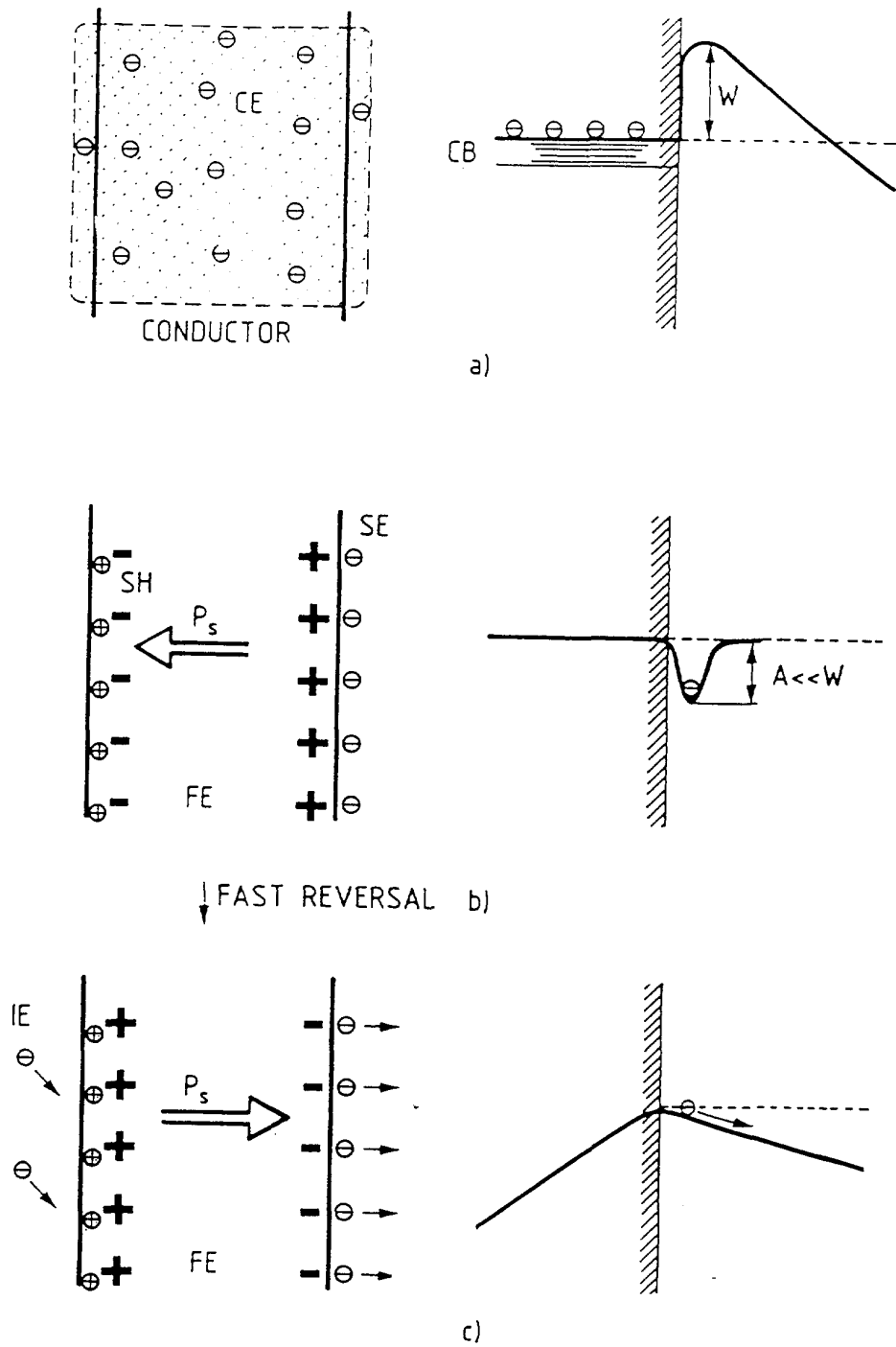


Fig. 1: Simplified principle of FE space charge emission after a rapid polarization change. a) Emission of conduction electrons from a metal ( $W$  = work function, CB = conduction band). b) Neutralization of FE by screening electrons (SE) and holes (SH). c) Emission from FE after  $P_s$  reversal and neutralization by injected electrons (IE).

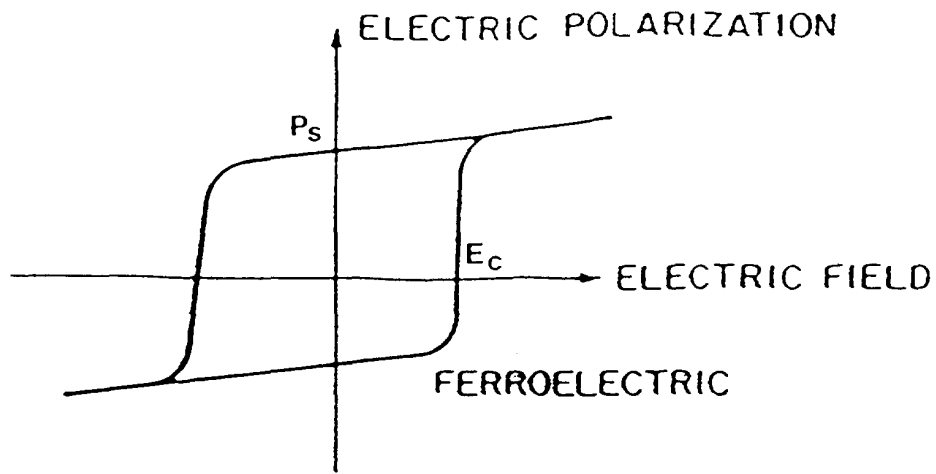


Fig. 2: Hysteresis loop of a typical FE material:  $P_s$  = spontaneous polarization,  $E_c$  = coercive field.

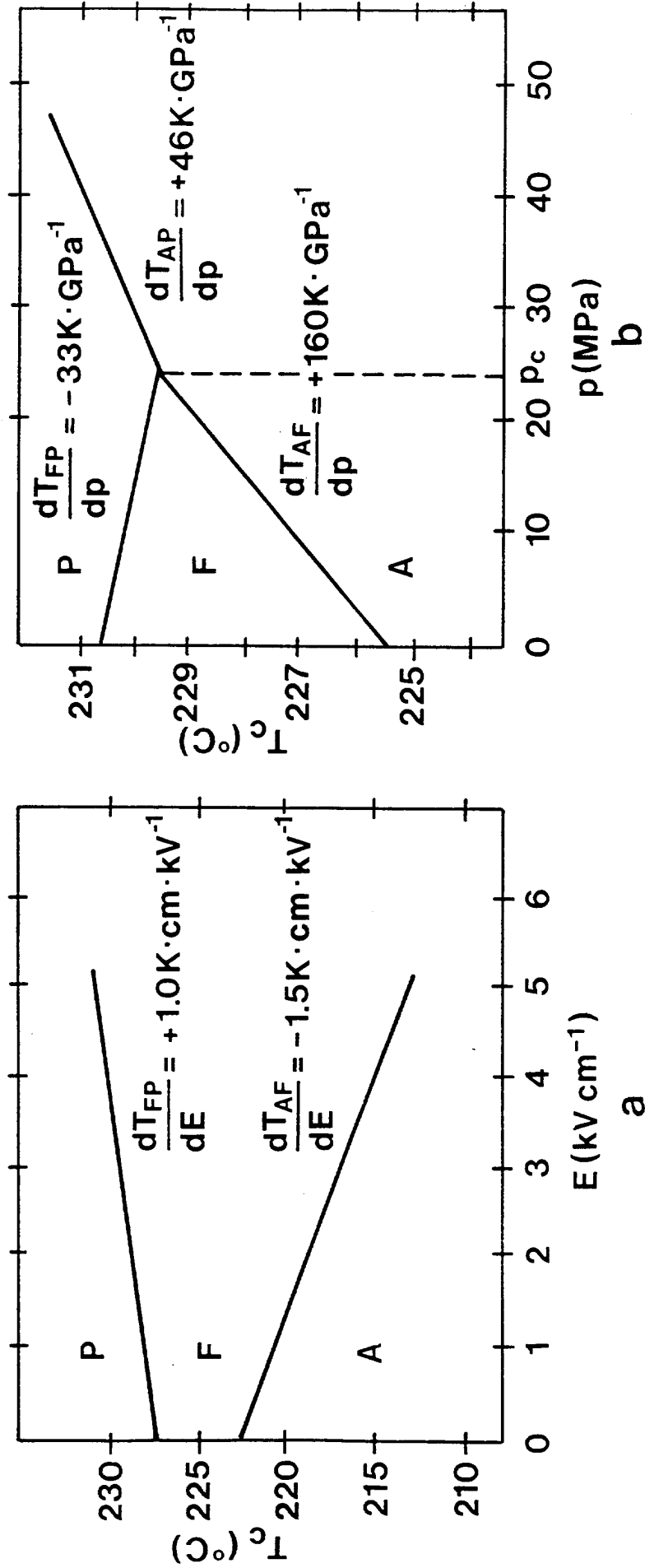


Fig. 3: Dependence of phase transition temperature  $T_c$  for antiferroelectric-ferroelectric (AF) and ferroelectric-paraelectric (FP) transitions on electrical (a) and mechanical (b) stress valid for PZT ceramics.

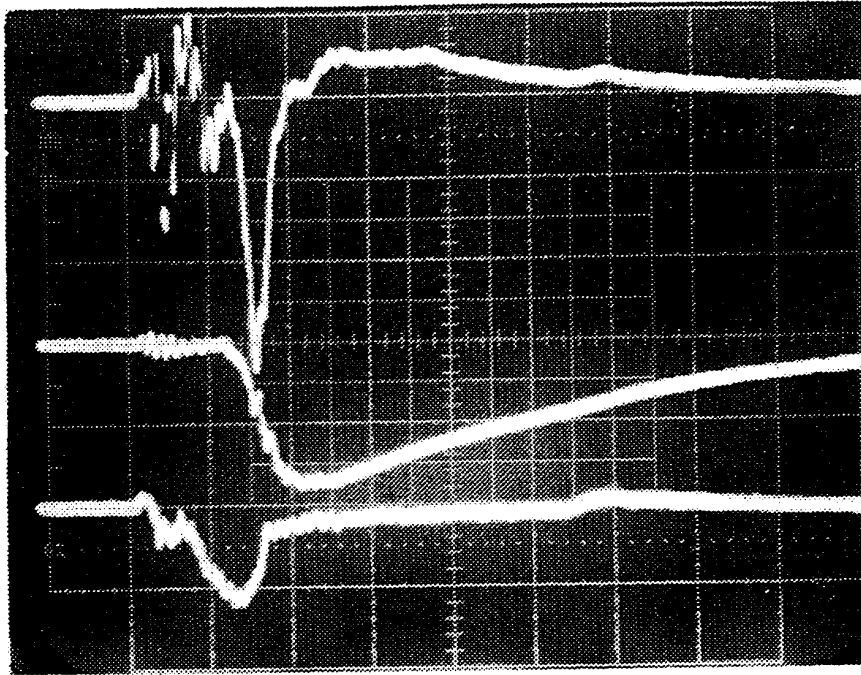


Fig. 4: Characteristic waveforms observed in real ferroelectric emission from a ceramic PLZT sample without external extraction voltage. Upper trace: emitted current ( $2 \text{ A/cm}^2$  per small division (sd),  $100 \text{ ns/sd}$ ). Middle trace: emitted charge ( $200 \text{ nC/sd}$ ). Lower trace: externally applied switching voltage ( $1 \text{ kV/sd}$ ).

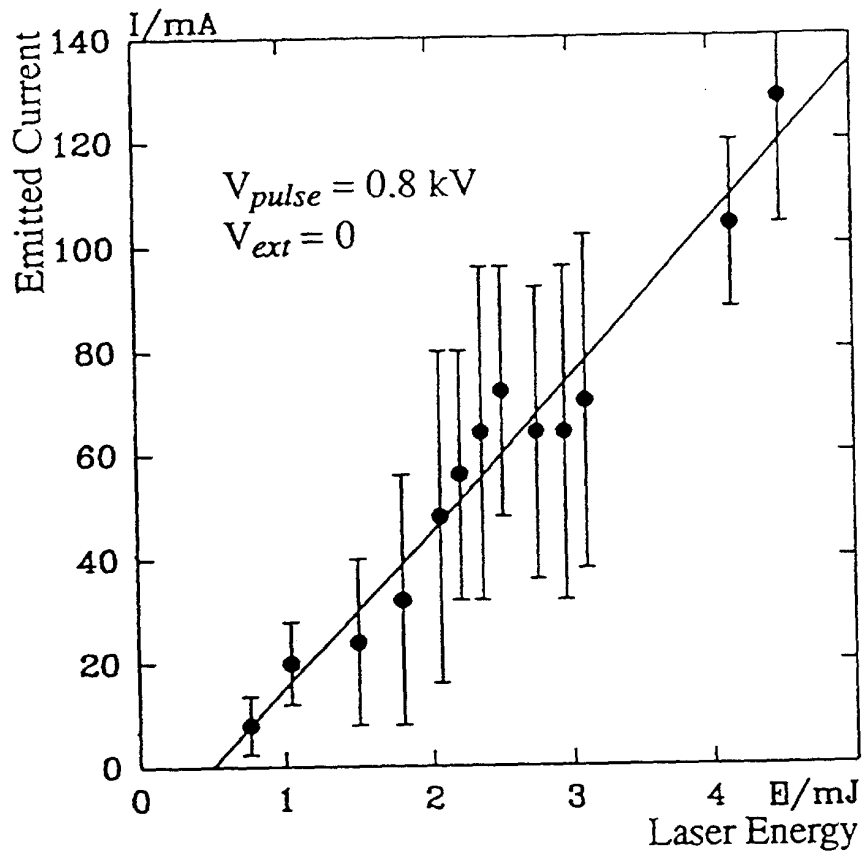


Fig. 5: Dependence of self-emitted electron-beam current on laser pulse energy at 355 nm wavelength from a PLZT 2/94.5/5.5 cathode. Threshold of laser pulse energy = 500  $\mu$ J. Previous  $P_s$  switching was done with a 0.8 kV high voltage pulse amplitude.

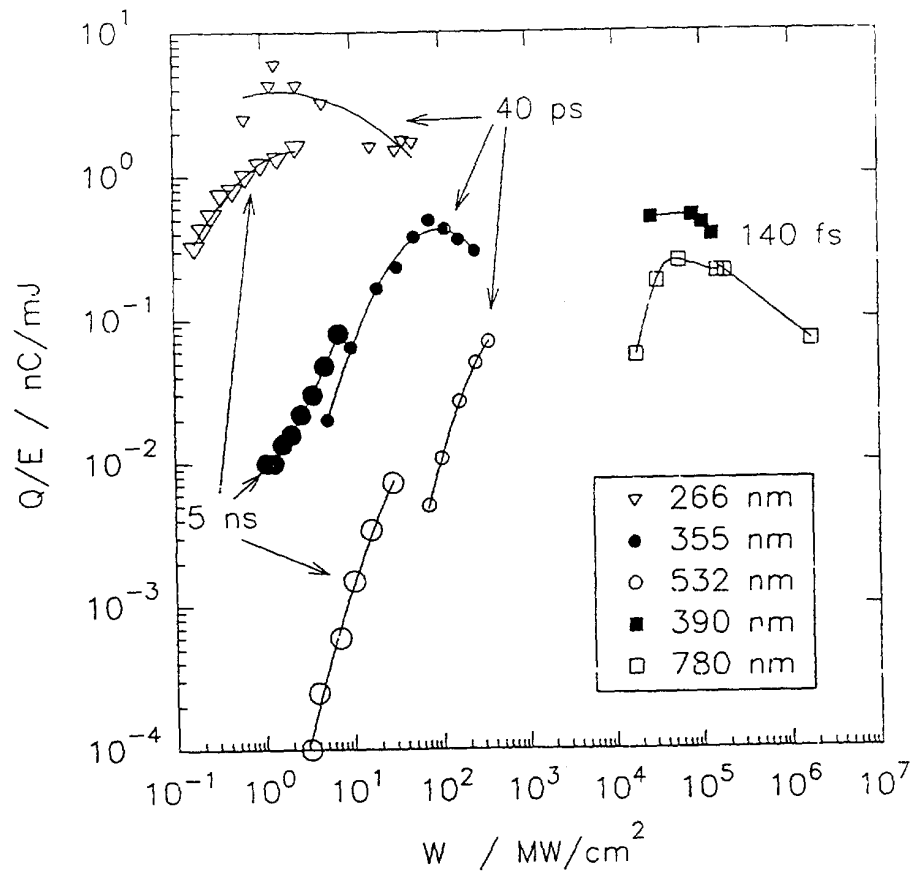


Fig. 6: Laser-induced emission efficiency of a PZT 95/5 pc doped with 2%  $Cr_2O_3$  for different laser wavelengths and for three laser pulse lengths (5 ns, 40 ps and 140 fs) as function of laser power density at 30 kV extraction voltage.

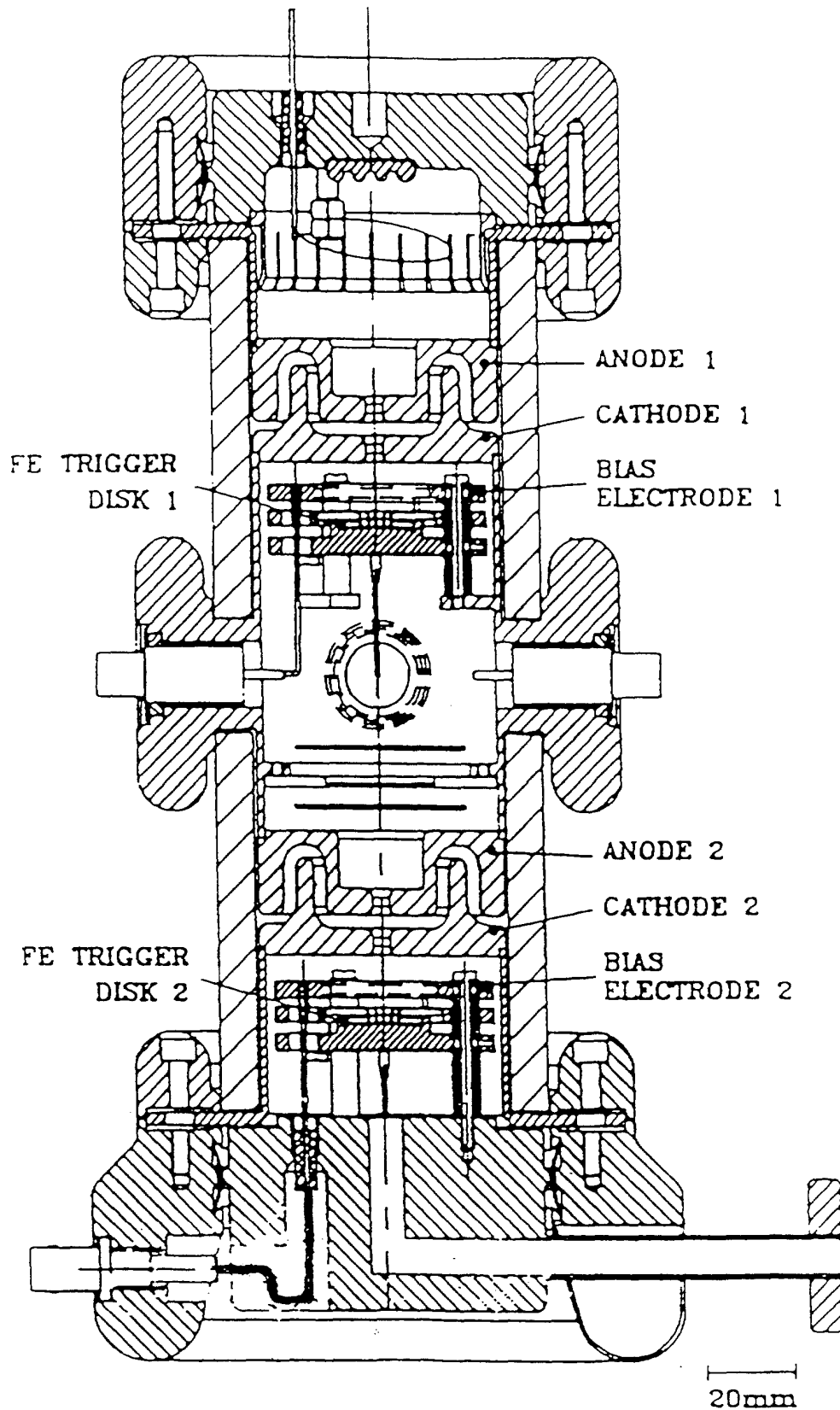
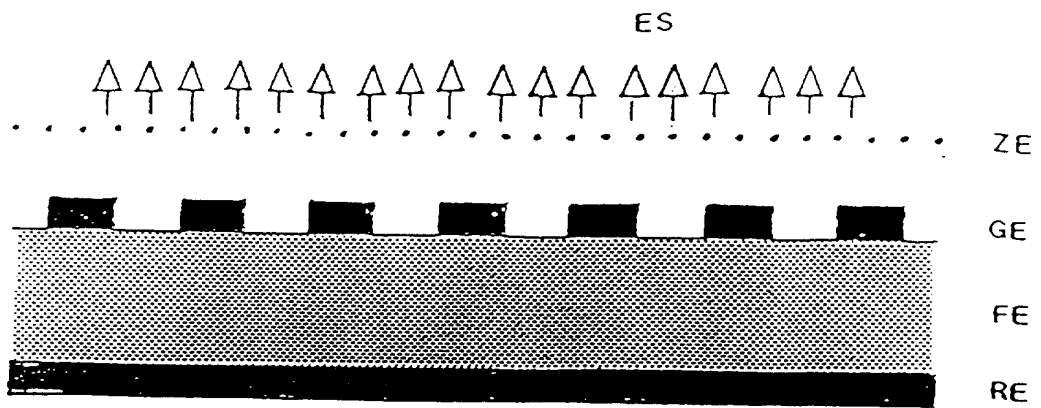
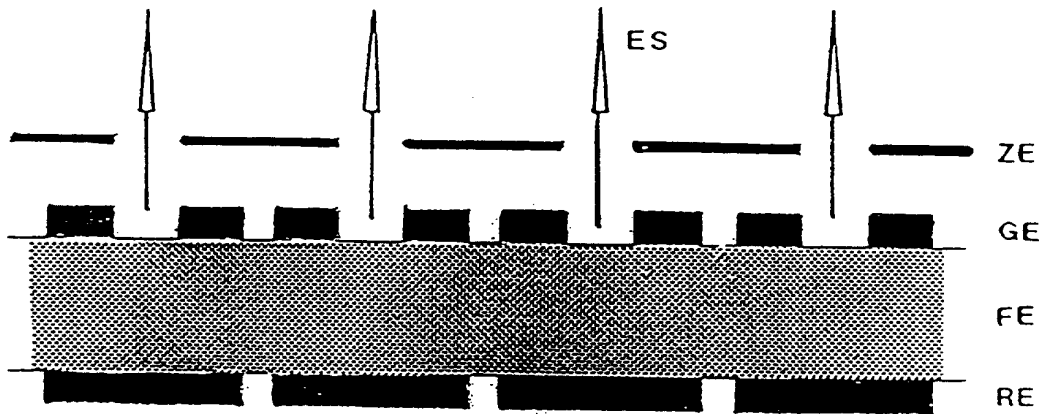


Fig. 7: Design of a two-gap pseudospark switch with two ferroelectric electron-beam triggers. Switch characteristics: maximum hold-off voltage = 30 kV; maximum current = 60 kA; switching precision = 10 ns; FE-trigger characteristics: beam current = 20 A gas-amplified five times; PLZI cathode thickness = 0.3 mm; free grid surface = 0.5 cm<sup>2</sup>.





a



b

Fig. 8: Scheme of thin-layer, large-area FFE emission, for the generation of a) large-area ribbon beams for microwave production, b) arrays of individual pixel-controlled electron beams for flat TV screens. EB = electron beam; ME = modulating electrode; GE = grid electrode; RE = rear electrode; FE = ferroelectric layer.

