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Electron Emission from Ferroelectrics - A Review

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Abstract

The strong pulsed emission of electrons from the surface of ferroelectric (FE) materials was discovered at CERN in 1987. Since then many aspects and properties of the method of generation and propagation of electron beams from FE have been studied experimentally. The method is based on macroscopic charge separation and self-emission of electrons under the influence of their own space-charge fields. Charge separation and electron emission can be achieved by rapid switching of the spontaneous, ferroelectric polarization. Polarization switching may be induced by application of electrical-field or mechanical-pressure pulses, as well as by thermal heating or laser illumination of the ferroelectric emitter. The most attractive features of FE emission are robustness and ease of manipulation of the emitter cathodes which can be transported through atmospheric air and used without any problems in vacuum, low-pressure gas or plasma environments. Large-area arrangements of multiple emitters, switched in interleaved mode, can produce electron beams of any shape, current amplitude or time structure. The application of the FE emission in accelerator technology has been demonstrated in several cases, e.g. for triggering high-power gas switches, for photocathodes and for electron-beam sources intended to generate, neutralize and enhance the yield of ion beams. Other applications can be envisaged in microwave power generators and in the fields of electronics and vacuum microelectronics[1].

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1. Introduction

In the early 6th century B. C. Thales of Miletus made the first observations of electricity by rubbing pieces of amber, which then attracted certain substances. This effect was visible thanks to the macroscopic charge separation, which was induced by the friction of the amber. Today everybody can easily observe space-charge emission between two pieces of dielectrics when separating the surfaces which were previously in contact. The same type of emission can also be realized with ferroelectric (FE) material in which charge separation can be achieved by changing the spontaneous ferroelectric polarization. In fact, almost one century ago, strong spark emission was produced in fire lighters by hitting a piece of FE material with a small hammer.

The electron emission during polarization reversal of ferroelectric 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. Current densities of the order of 10^{-12} A/cm^2 were quoted. In 1987 Gundel, Handerek, Riege and Zioutas were able to produce "strong" electron emission from FE materials with current densities of more than 100 A/cm^2 in the absence of any external extraction field. This strong electron emission is excited by submicrosecond polarization switching with high-voltage pulses [3–7] applied via 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. The self-emission of high current densities indicates that the electrons originate after switching from the negatively charged surface. The emission is enhanced by the electrons which have screened a large part of the positive spontaneous polarization charge before switching. The ferroelectric emission, hence, very much resembles the spark emission from dielectric surfaces induced by friction and subsequent charge separation. Recently it has been shown at CERN that laser-induced electron emission from FE materials is based on spontaneous polarization switching just as field-excited and pressure-induced FE emission, [8], and therefore differs strongly from classical photoemission.

Meanwhile, research and development on FE emission and related applications has been started or is continuing in several countries all over the world [9–16]. The purpose of this paper is not so much to present the role of microscopic ferroelectric physics during the emission, as to describe phenomenologically the processes of emission and beam formation starting at the surface under the influence of the charge distributions and the related space-charge and polarization fields. Furthermore, the implications of ferroelectric emission for practical applications are pointed out and examples of successful developments in accelerator technology are presented.

2. Principles of Ferroelectric Emission

2.1 Simplified mechanism

The main difference between the classical methods of electron emission (thermionic, field, secondary and photoemission) and FE emission is the degree of charge separation before emission occurs. In addition to raising the energy level locally by heat, by photons or by secondary particles, which liberate electrons from the lattice, the conventional methods require high extraction potentials in order to extract the charges from the neutral surface. Fast polarization switching is the trick to generate a macroscopic charge separation on the two opposite surfaces of an FE sample. The resulting space-charge (polarization) fields are so high that self-emission takes place.

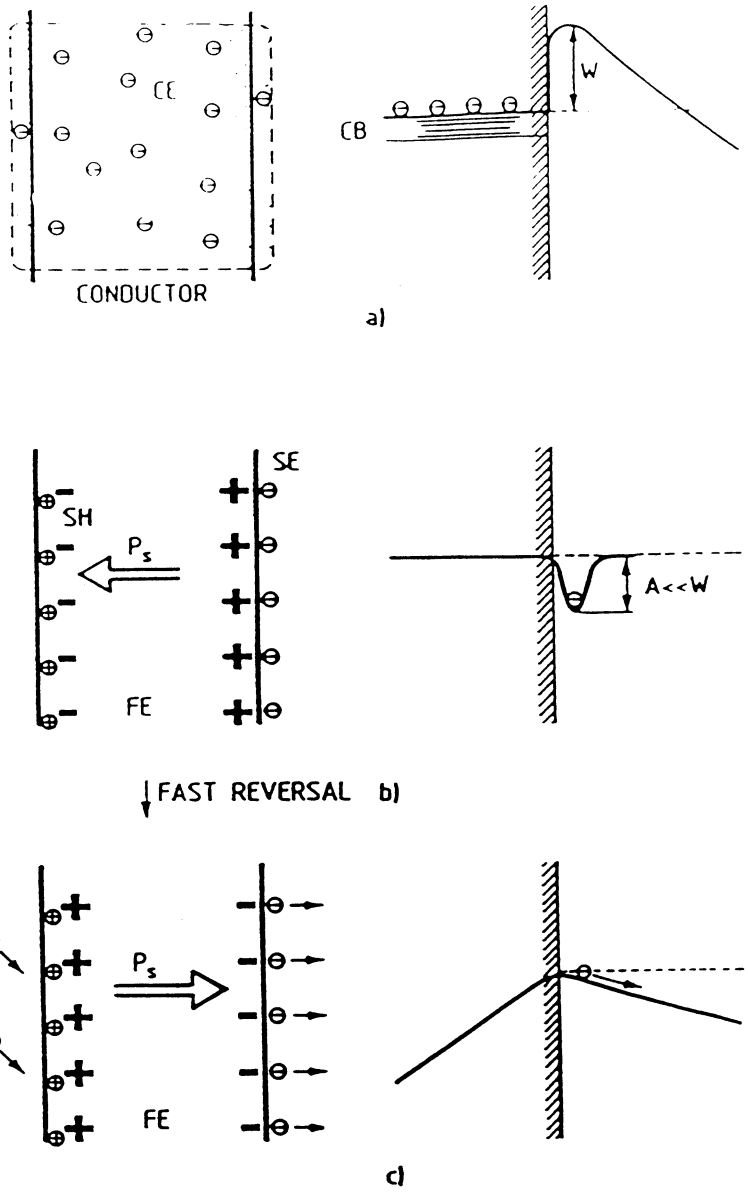


Figure 1: Principle of FE space charge emission after a polarization change. a) Emission of conduction electrons from a metal (W = work function, CB = conduction band). b) Neutralization of FE by screening electrons (SE) and holes (SH). c) Emission from FE after P_s reversal and neutralization by injected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

Figure 1 gives a simplified idea of the emission process: the FE sample is represented by a single-domain structure with two possible directions of the spontaneous polarization vector P_s. In reality,

even in FE single crystals, a multi-domain structure is present with several possible orientations of P_s. 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. 2a. For multi-grain and multi-domain materials an average hysteresis curve can be defined and measured, but emission can take place from any domain at any surface where the necessary electric field conditions are reached. In fig. 2b P_s is plotted schematically as a function of temperature. At the transition temperature from the FE to a non-ferroelectric phase P_s decays to zero. By prepoling an FE sample at higher temperatures under the influence of a modest electric d.c. field the hysteresis curve can be shifted with respect to the former, symmetric origin [17]. A threshold field of the order of the coercive field strength E_c has to be applied before emission starts. It is, however, important to raise the field in a very short time to the switching amplitude, otherwise the charges produced by the statistical domain switching can flow away over the surface or through the bulk of the FE material.

2.2 Methods and features of FE emission

Fast switching of spontaneous polarization can be achieved in several ways. It is not necessary to completely reverse P_s as indicated in figs. 1 and 2, but to change only a part ΔP_s of P_s either by moving under electric-field application along the hysteresis curve (fig. 2a), by creating or annihilating FE domains through temperature changes (fig. 2b), or by crossing phase boundaries under application of electric-field or mechanical-pressure pulses (fig. 3a and b). Negative surface charges are always formed on the surface where the positive polarization charge is decreased.

Figures 4a and 4b show the emission of a current density of 10^8 electrons per cm^2 with kinetic energies of 25 keV from a triglycinesulfate (TGS) single crystal, which was placed in a β spectrometer of high resolution and heated slowly across the phase transition temperature at 50^0C [3]. Emission is observed in spite of the slow heating rate, because of the sporadic, but fast switching of individual domains from the FE to the non-ferroelectric state. This process can be compared to the Barkhausen effect in ferromagnetic materials. When the same heating test is performed in a microwave resonator cavity, microwave bursts, which are also caused by phase transition switching of individual domains, are observed at frequencies in the GHz range.

Figure 5 shows P_s switching by inducing fast phase transitions with high-voltage pulses applied to a lead-zirconium-titanate (PZT) ceramic sample with a titanium content of 5 % and with a phase diagram corresponding to fig. 3a. The upper phase limit between the so-called intermediate FE phase and the paraelectric (PE) phase rises with applied field amplitude, whereas the lower boundary between the FE and the antiferroelectric (AFE) phase decays with electric field. In the same manner, switching and electron emission can be induced by fast phase transitions with mechanical pressure pulses (fig. 3b) as is known from the old-fashioned fire lighters.

Polarization switching, induced by the application of HV pulses to disk-shaped FE samples via gridded electrodes, has been applied in most of the emission experiments that have been carried out since the discovery of strong FE emission. Figure 6 shows typical emitted current and charge waveforms induced by a negative HV pulse applied to the rear electrode of an FE sample previously prepoled with a negative d.c. voltage applied to the grid electrode. More complicated patterns of field-excited electron emission (FEEE) may be observed depending on the type and prepoling of the FE material as well on the electrical excitation and discharge circuit parameters. In fully ferroelectric material the emission is mainly bound to P_s reversal processes, whereas in materials composed of different phases (e.g. FE materials near a diffuse phase boundary) fast transition processes from one phase to another may play a major role. When optimized emission with minimum mechanical and electrical fatigue is aimed at, the main technological problems of FEEE to be mastered are the choice of composition, the production and the preparation of the material, the processes of electroding and insulating the FE cathode surfaces, and the prepoling procedure.

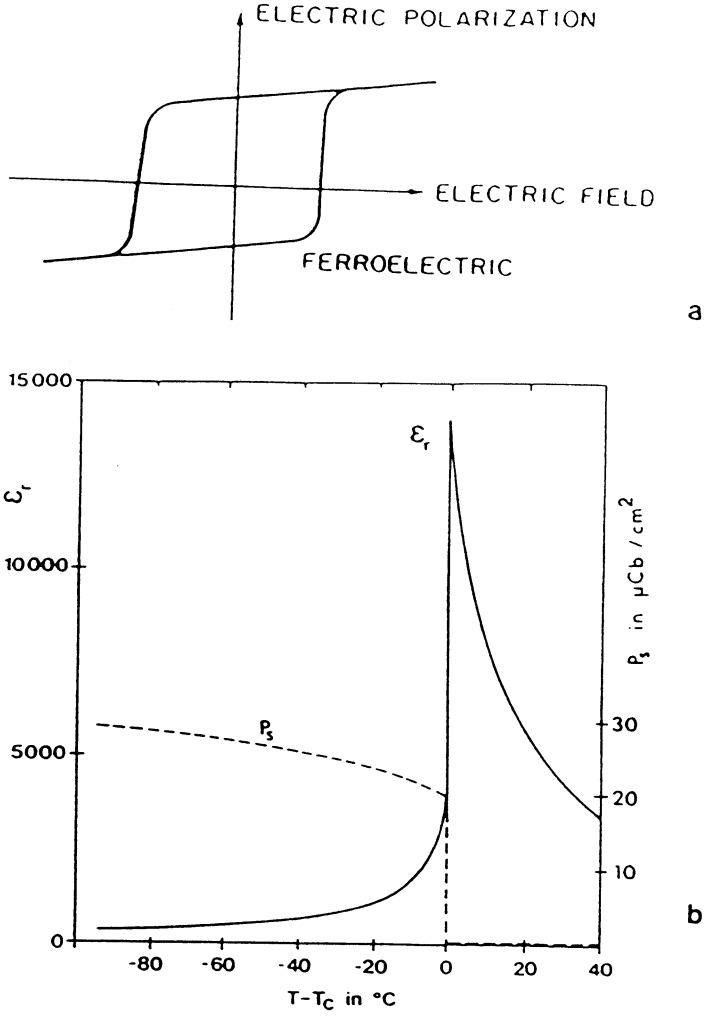


Figure 2: a) Hysteresis loop of a typical FE material: P_s = spontaneous polarization, E_c = coercive field. b) Temperature dependence of P_s and dielectric constant ε_r of an ideal FE: T_c = phase transition temperature.

The HV switching pulses needed for lead-lanthanum-zirconium-titanate (PLZT) samples of 1 mm thickness usually have amplitudes greater than 1 kV and pulse lengths ranging from 100 to 1000 ns. It has been shown that the HV switching voltage amplitude can be decreased according to FE sample thickness. The lowest pulse voltage reached so far by switching 30 – 45 µm thick PZT samples was 75 V [12]. Under these conditions current densities of the order of several A/cm^2 were emitted. 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 1 – 100 nF/cm^2 depending on thickness and material, high-current switching pulse amplitudes of the order of $10^2 - 10^3$ A are needed when beams with large cross-sections have to be generated. In many cases the conversion efficiency from primary electric

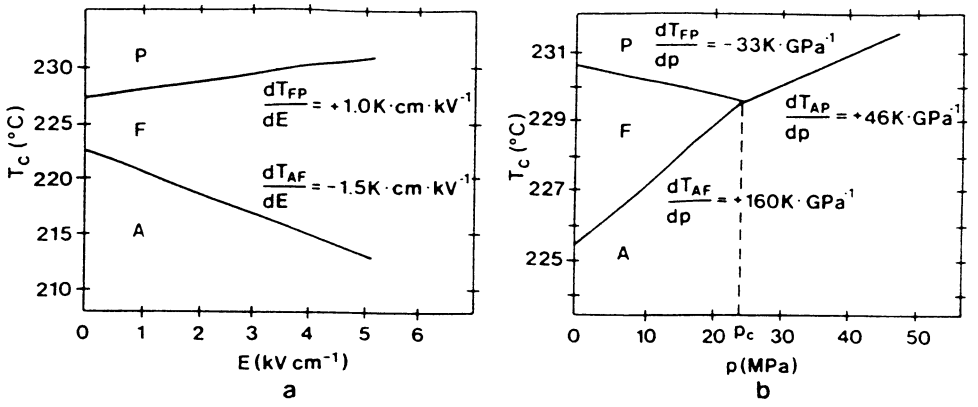


Figure 3: Dependence of phase transition temperature T_c for antiferroelectric-ferroelectric (AF) and ferroelectric-paraelectric (FP) transitions on electrical (a) and mechanical (b) stress valid for PZT ceramics.

switching energy into electron beam energy has exceeded 10%. Emitted current densities with amplitudes above 100 A/cm^2 and pulse lengths shorter than the excitation pulse length, but strongly dependent on the FE material type, have been measured. The emission is independent of the quality of the vacuum, but can be strongly enhanced in the presence of a low-pressure gas or of a plasma [18]. A question which is often raised refers to the limitations of the emitted current density. Apparently the total emitted charge density per pulse is given by the material constant P_s (for $LiNbO_3$, $P_s = 1$ C/m^2 = 100 μC/cm^2). The emitted current density depends on the rate of P_s change [6]. In practice, however, no upper limit is observed. The reason is the formation and spreading of a plasma 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 FEEE, can be far below 100 A/cm^2. As is known, plasma-assisted emission is unlimited with respect to current density and independent of the emitting material. Strong plasma formation can have negative consequences, especially in the presence of high d.c. or a.c. extraction potentials. Even in vacuum a total electrical breakdown may be ignited. A weak surface plasma, on the other hand, has several beneficial effects, such as space-charge elimination (favourable for beam emittance), fast recharging of the unscreened FE surface after switching, and homogenization of the emission.

A maximum repetition frequency of 2 MHz limited by the HV pulse generator has been obtained with FEEE [5]. Repeated pulsing works without the application of a reset pulse. Rapid recharging, most probably via the aforementioned surface plasma, and fast repolarization turns the sample back to its original state (fig. 7). In most cases the emission starts above a threshold of the excitation field, which is only weakly dependent on the influence of an additional extraction potential (fig. 8a), and continues to rise more than linearly with rising HV pulse amplitude. A similar characteristic is observed for the dependence of emitted charge on extraction field (fig. 8b).

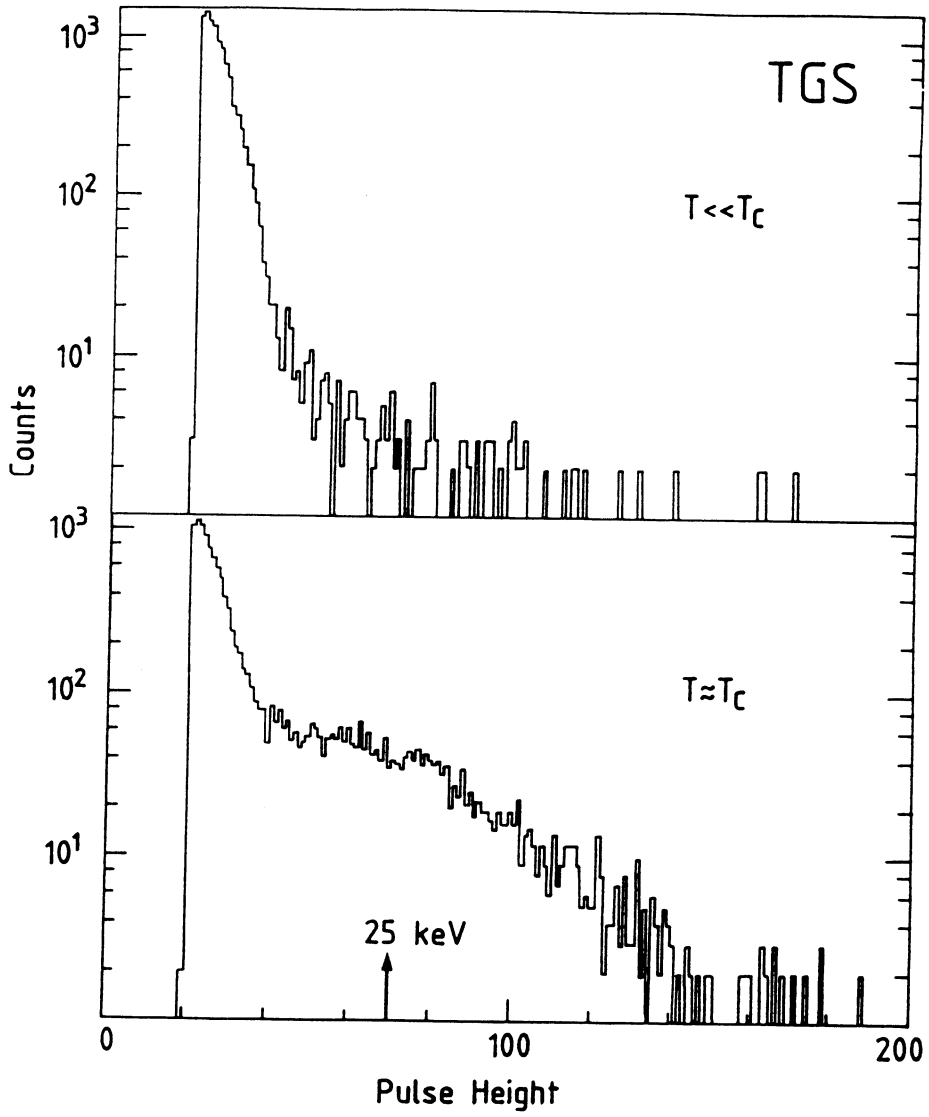


Figure 4: Pulse-height spectrum of electrons measured with a β spectrometer for two temperatures of a TGS crystal (5 mm diameter, and 3 mm thickness) during heating. Near T_c (50° C) the measured kinetic electron energy was 25 keV (a). The PM background at a room temperature is shown in (b).

2.3 Laser-induced electron emission from FE

Until recently laser-induced emission from FE (LIEE) was considered to be different from the previously described FE emission methods. Since the emitted electron beam current and the light-intensity pulse had the same shape, LIEE was believed to be a special case of conventional photoemission with the difference that the polarization screening electrons would be the emitted ones and not the electrons from the bulk of the FE material [19 – 20]. This model had to be abandoned when strong self-emission of energetic (> 10 keV) electrons by laser irradiation of FE cathodes was observed without any extraction field (Fig. 9 and [8], [21]). Kinetic electron energies of this order of magnitude can be explained only by macroscopic charge separation in the FE sample induced by polarization

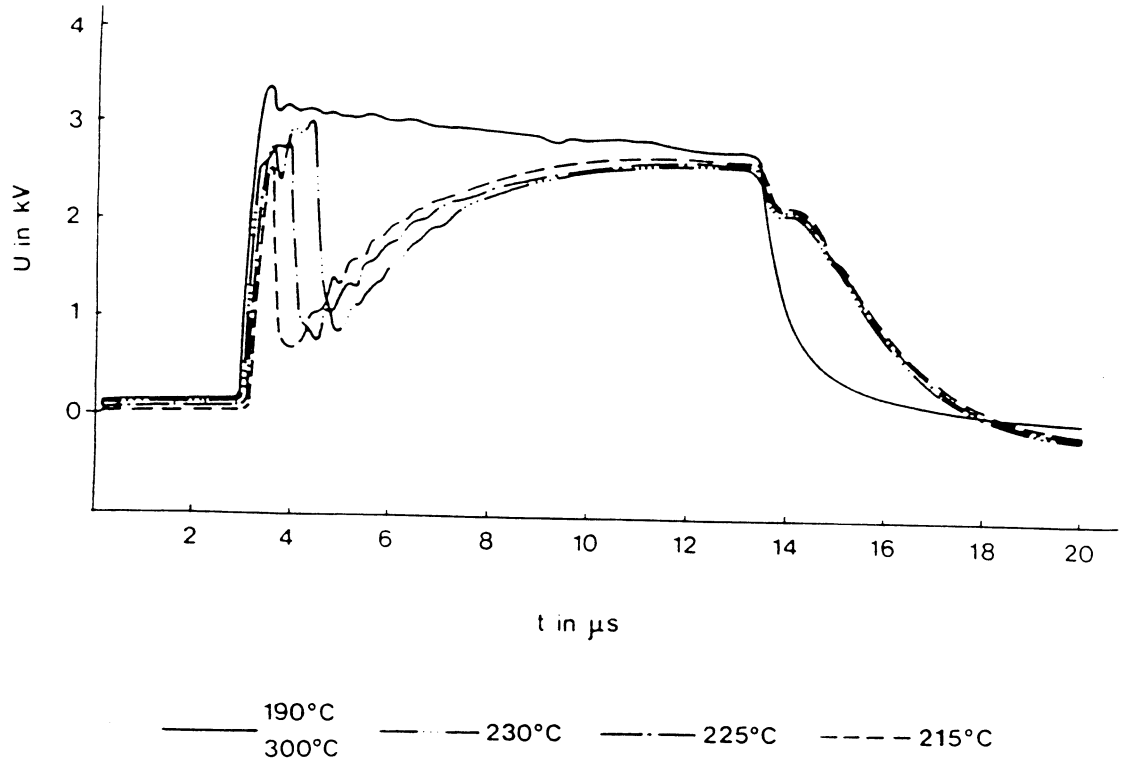


Figure 5: Fast phase transition switching (waveforms at 215^0 C and 230^0 C) across the AFE-FE and the FE-PE phase boundaries can be achieved via a horizontal crossing in the temperature-field diagram (Fig. 3a). Inside the FE state (waveform at 225^0 C), P_s is reversed. Outside the FE state (300^0 C and 190^0 C) there is no indication of a polarization change (rectangular pulse form).

switching through the laser pulse. LIEE therefore resembles FEEE and is totally different from classical photoemission.

The strength of LIEE can be influenced by several factors, such as by the prepoling of the FE cathode, by previous polarization switching with HV pulses, or by laser irradiation without extraction voltage. Enhancement of normal LIEE up to a factor of 200 was observed under certain conditions [20]. The enhancement factors decrease with rising extraction field. Self-emission also requires regular polarization switching in order to be maintained at a constant emission level. Without such refreshment the self-emission decays after some tens of LIEE pulses towards zero.

At nanosecond timescale no time-lag could be observed between laser and electron-beam pulse. This proves that polarization switching of FE material can be achieved in less than 1 ns. Still another type of polarization switching by laser irradiation was observed (fig. 10): with a ceramic PLZT 1/94/6 cathode total (bulk) polarization reversal could be reproducibly initiated with a single laser pulse in the absence of any HV pulse switching. The temporal evolution of the corresponding emission is much slower than the LIEE and independent of the laser pulse shape. On the other hand, the amount of switchable P_s available in LIEE experiments was less than 0.1 % of the total P_s with the FE material used up to now. The parameters that determine the percentage of LIEE-switchable

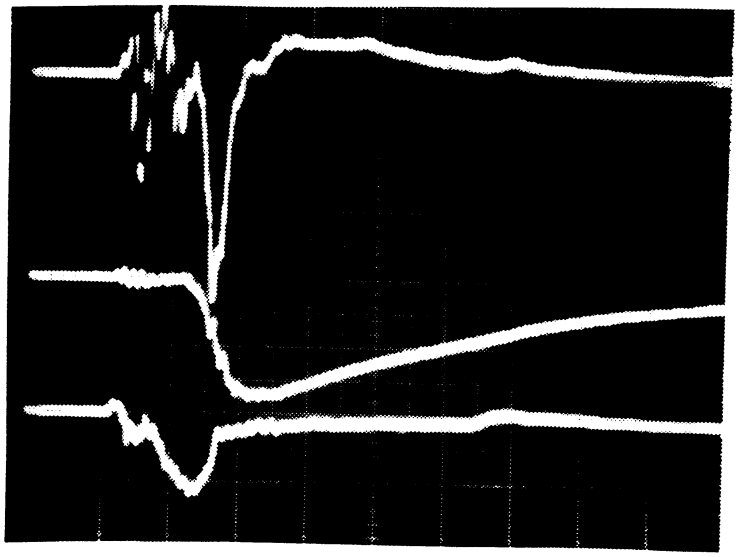


Figure 6: Typical waveforms of emitted current (upper trace, 2 A/cm² per small division (sd), 100 ns/sd), charge (middle, 200 nC/sd), and switching voltage (lower trace, 1 kV/sd) obtained from a ceramic PLZT sample without extraction voltage.

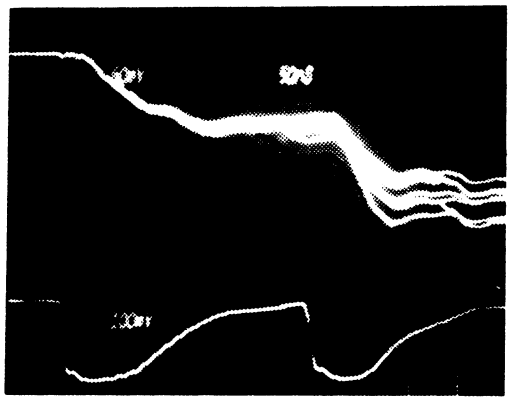
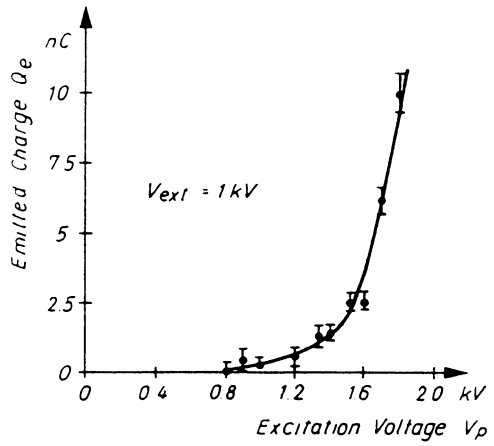
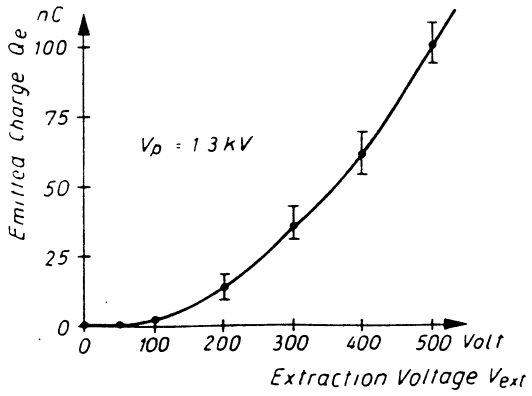


Figure 7: FEEE emission at 2 MHz repetition rate from a PLZT 2/95/5 sample of 1 mm thickness. Top: emitted charge, 5 nC/sd, 50 ns/sd. Bottom: HV pulse (1 kV/sd).

P_s, are FE material phase composition, prepoling of the cathode, absorption coefficient of the FE material, roughness of emitting surface, interaction with FEEE operation, presence of extraction voltage, level of laser-pulse energy, and wavelength. In the same way as for FEEE and contrary to classical



a

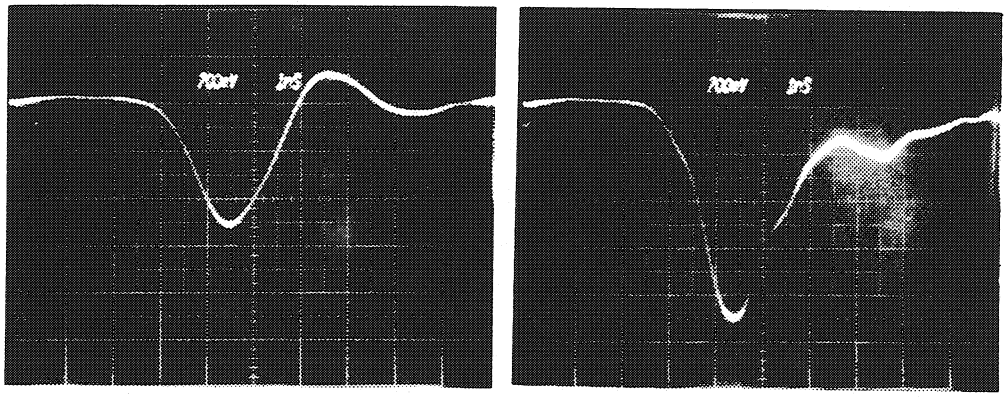


b

Figure 8: Emitted charge from a lanthanum (1% La_2O_3) doped (Ba,Sr)TiO_3 solid solution sample as a function of HV excitation pulse voltage (a) and of extraction voltage V_{ext} (b) with an emitting area of 4 mm^2.

photoemission, a threshold laser energy is observed, below which emission stops (fig. 11). For the case of self-emission a conversion efficiency of laser into electron-beam energy of less than 3% was calculated not taking into account the polarization switching energy needed for the occasional refreshment of the self-emission process.

The conversion efficiency in the case of self-emission can be measured easily, but not in a real electron gun, where high extraction fields are always needed to transport the electron beam away. The term of quantum efficiency (QE) can be used artificially for comparing in practice FE with standard cathodes. Physically, however, it is meaningless, since LIEE is not limited primarily by potential barriers and should function with electromagnetic radiation of any wavelength, provided the energy is absorbed in the surface layer of the FE cathode. For practical applications in electron sources the emitted charge per unit of laser energy (e.g. C/J or nC/μJ) can serve as a convenient definition of efficiency.



a b

Figure 9: a) Waveform of self-emitted electron beam current from PLZT 2/94.5/5.5 at 2.2 mJ laser pulse energy and 355 nm wavelength measured just after polarization switching with a HV pulse amplitude of 1.5 kV and 700 ns duration. b) Waveform of emitted electron beam current measured immediately after a polarization switching under 8 kV extraction voltage voltage from the same sample and with the same laser parameters. (16 mA/sd, 1 ns/sd)

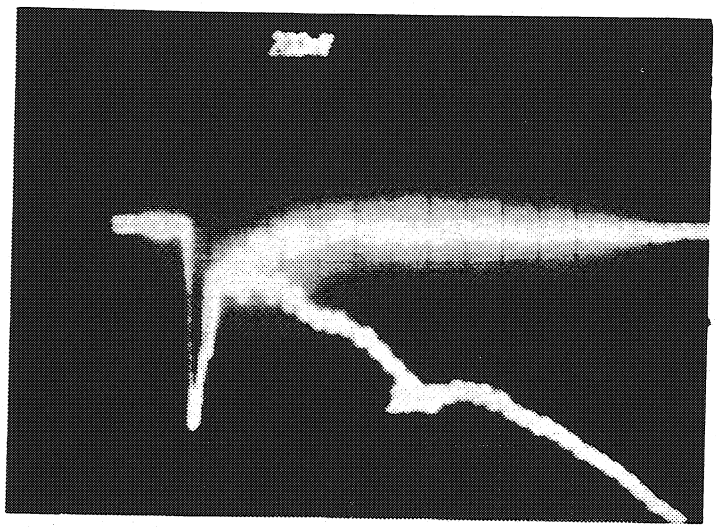


Figure 10: Total P_s reversal of a 1/94/6 PLZT cathode induced entirely by a single laser pulse in the absence of HV switching, but with an extraction potential of 6 kV.

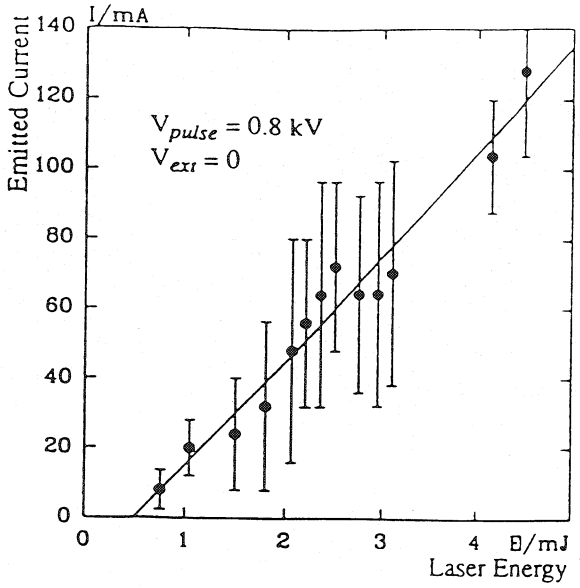


Figure 11: Dependence of self-emitted electron-beam current on laser pulse energy at 355 nm wavelength from a PLZT 2/94.5/5.5 cathode. Threshold of laser pulse energy = 500 μJ. Previous P_s switching was done with a 0.8 kV HV pulse amplitude.

3. Applications

3.1 Basic advantages of FE cathodes for practical applications

The main advantage of FE cathodes in technical applications [22] is certainly not the high intensity of electron emission, in the first line; for most cases considered up to now the necessary charges and currents could easily be produced, however, more important is the ease of practical use. The very robust ceramic PLZT material compares very well in reliability with the best metallic cathodes, while producing much stronger emission with FEEE, as well as with LIEE. FE cathodes can be touched without any consequences on the surface, transported through air without care, and do not need good vacuum for operation. They work in (low-pressure) gas and plasma. Especially in high-power, cw applications, the features of good conversion efficiency of primary into beam energy, as well as the high-repetition-rate performance of certain FE ceramics 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, so that the Langmuir-Child limitation can be exceeded by several orders of magnitude [10]. For LIEE at long wavelengths, appropriate materials may be found which soften the operating conditions for the laser systems and avoid the generation by stray light of parasitic electrons in the metallic source cavities.

3.2 Applications in accelerator technology

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 reliabilty, robustness and emitted current densities, more flexibility in spatial and temporal beam pulse shaping, and easier transportability are the main technical advantages. A typical example is the high bunch charge version of an electron gun for the linear collider injector of a superconducting, TESLA-type accelerator [23], which requires 100 A, 10 ps electron-beam pulses with a repetition rate of 1 MHz. These requirements could be reached, at least up to a charge level of 20%, with simple FEEE-driven FE cathodes. Other applications in the accelerator area are electron sources for large-volume gas ionization or large-area surface irradiation, e.g. for x-ray generation.

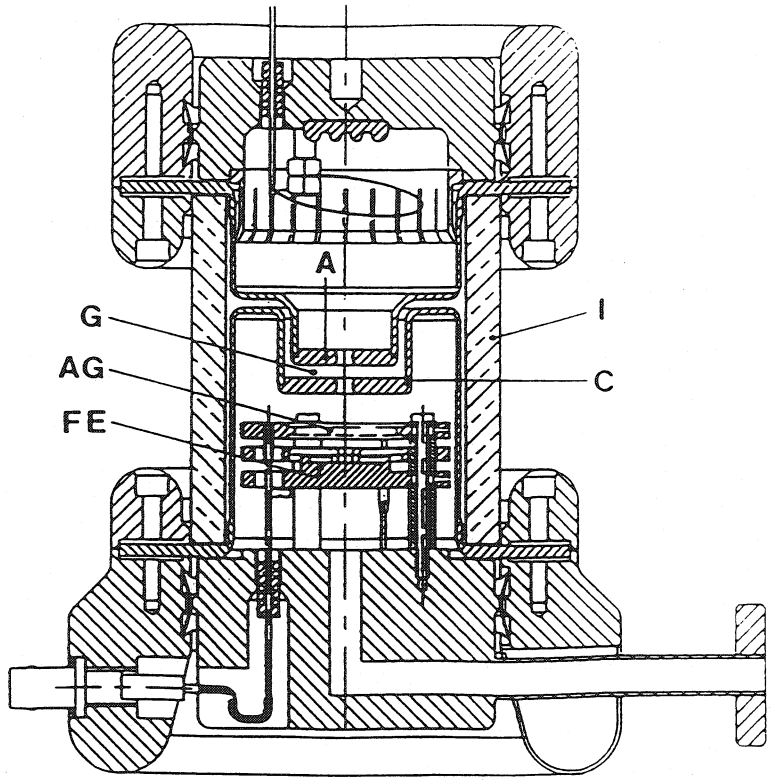


Figure 12: Design of a one-gap pseudospark switch with ferroelectric electron-beam trigger. Switch characteristics: maximum hold-off voltage = 30 kV; maximum current = 45 kA; switching precision = 1 ns; time between erratic breakdowns > 10^3 hours. FE-trigger characteristics: beam current = 20 A, gas-amplified five times; PLZT 2/95/5 cathode thickness = 0.3 mm; free grid surface = 0.8 cm^2.

One example for such a dedicated source is the electron-beam trigger system of the high-power, low-pressure pseudospark switches of the pulse generator for the future LHC beam dumping system [24 – 26]. The FE cathode emits an annular beam of 10 – 20 A (60 A/cm^2), which is amplified several times by the presence of a low extraction field (300 V) and of a low-pressure deuterium atmosphere.

The beam is sent from the interior of the main hollow cathode into the main gap region of the switch and initiates, with nanosecond precision, a 30 kA discharge of the storage capacitor charged to a voltage in the range 1.5 – 35 kV (Fig. 12). The switching and emitting efficiency and the long-term reliability of the FE cathode under the very difficult environmental conditions of contact with hot plasma and metal vapor, have proved to be excellent in many prototype tests. Over a period of several years not a single cathode was destroyed and no missing trigger pulse observed. Industrial production of FE trigger cathodes has been started.

Another application of FE emitters are short-pulse (several ps), laser-driven FE photocathodes. At wavelengths of up to 350 nm nC charges contained in a 5 ns pulse length have been reliably and reproducibly generated during several 10^6 shots ([8] and [27 – 28]). Higher charge populations cannot be dealt with in today's accelerator injectors. Of real interest will be the development of laser-driven FE cathodes at wavelengths in the visible or infrared range. The LIEE emission mechanism is governed by the amount of energy absorbed in the cathode surface layer and not by potential barriers linked to the lattice structure of the material. Hence, even microwaves should provoke LIEE emission, provided the absorbed energy density is sufficiently high. The light sources, including lasers, become less problematic and more stable at longer wavelengths since frequency multiplication will be obsolete. Light reflected from the cathode cannot liberate from the walls of the source cavity the parasitic electrons which generally disturb the normal operation of the source. There are several factors, which shift the absorption edge of FE material towards longer wavelengths, e.g. material composition and doping [29], difference between working and phase transition temperatures, prepoling, internal electric fields, and surface roughness.

Experimentally it has been shown that the electron beams emitted from FE by FEEE can be used to induce a diffuse vacuum discharge and to sputter ions from a target to form a strong ion-beam source [30]. This procedure works especially efficiently, when some kind of ion-beam neutralization is applied. In reference [31] 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 electron beam sent from the outside into the source [32]. Such FE-generated beam neutralization could be used in future high-current ion linacs, e.g. in heavy-ion fusion drivers, to transport the required beam currents in a small number of accelerator pipes and to succeed in beam collision at the fusion target without space-charge blow-up. In addition to the power consumed by the ion driver linac, peak electron-beam generation powers of several GW and average powers in the MW range would have to be taken into account in the break-even conditions.

High-gradient particle acceleration (e.g. in a switched-power type of linac) was the starting point for the discovery of FE electron emission and is still of interest. The technological developments which have to be envisaged for this purpose are estimated however, to be much more difficult and the costs much higher than those required for FE emission developments.

Finally FE cathodes could serve as high-energy particle detector elements with picosecond response. A proof-of-principle experiment is under way in which the amount of polarization change and consequently the number of liberated electrons from an FE cathode by an energetic particle will be measured.

3.3 Industrial applications

Though the FE emission development work at CERN is mainly directed towards accelerator applications, FE electron emission can be and is already applied in a growing number of other technical fields. Interest has been raised in the use of FE cathodes in high-power tubes, such as klystrons or gyratrons, which would profit from higher current densities 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.

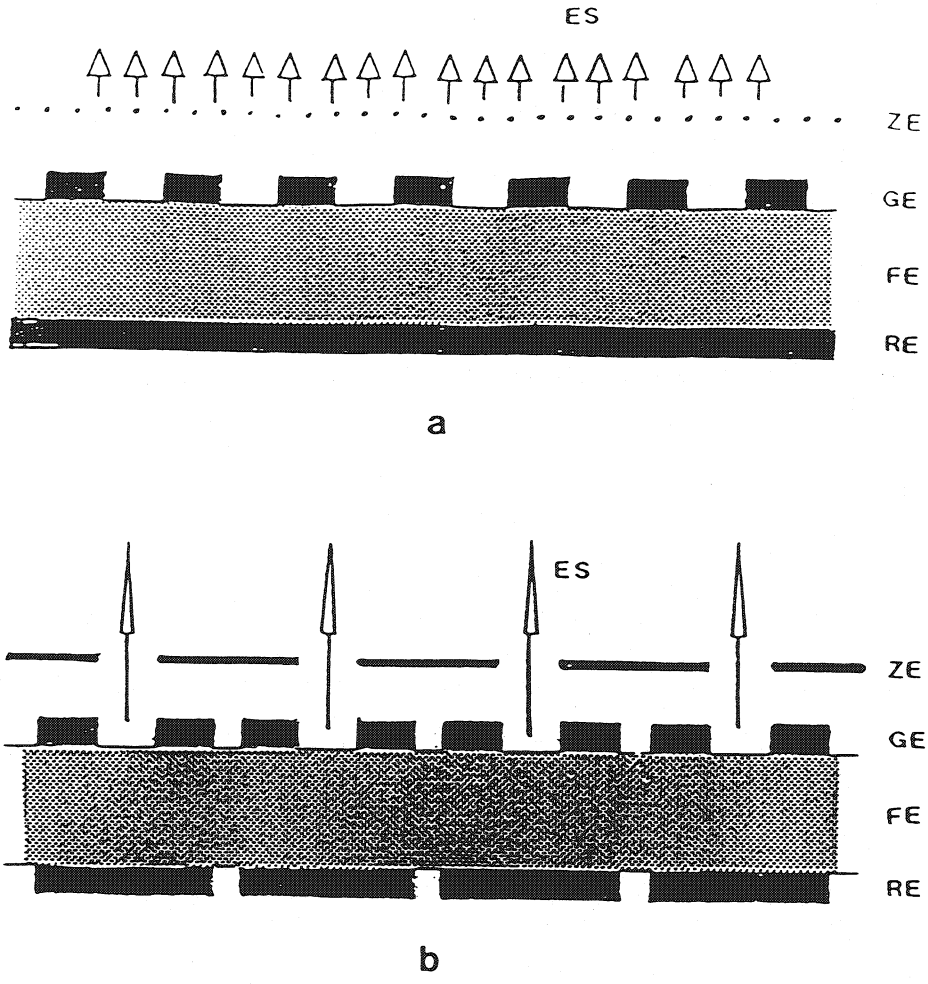


Figure 13: Scheme of thin-layer, large-area FEEE emission for the generation of a) large-area ribbon beams for microwave production, b) arrays of individual pixel-controlled electron beams for flat TV screens. EB = electron beam; ME = modulating electrode; GE = grid electrode; RE = rear electrode; FE = ferroelectric layer.

The largest number of applications can be imagined in electronics, microelectronics and vacuum-microelectronics. Components producing microscopic electron beams by FEEE and/or combinations with multi-microlaser arrangements (LIEE) and nanotechniques may lead to revolutionary new

devices. Work has already started in institutes and industry on the development of ultraflat TV screens using FEEE from thin FE layers (fig. 13). The same technique could be applied to the generation of GHz power from large-area thin FE layers as was proposed earlier with laser-driven, ribbon-like photocathodes (lasertron) or with large micro-tip arrays, which produce electron beams by field emission. [33 – 34].

4. Conclusions

The discovery of strong electron emission from switched FE material, which was considered a curiosity a few years ago, is now attracting increasing attention. The simple way to reliably generate electrons and the robustness of the ceramic FE material together have led to some technical applications and have triggered developments in the field of particle sources for accelerators, in the field of pulsed-power and power generation technology, and in electronics and vacuum-microelectronics. The author believes that this is just the beginning of a wider expansion into various fields of electron-beam devices. In many applications where high-power electron beams are involved, FE cathodes could replace thermionic or field emission cathodes thanks to the capabilities of high current density emission and long operational performance. New electron-beam devices can be designed in electronics and vacuum-microelectronics in combination with microlaser and nanotechnical systems. Technology transfer to industry seems fast in this particular case, since many techniques required for producing FE-emitting devices, such as production methods of ceramic thick-film and thin-film layers, are wellknown and available in the electronics industry. Nevertheless, a major development effort is required for most new applications where the generation of optimum emission under reliable conditions is envisaged.

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