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**FEMTOSECOND LASER-INDUCED ELECTRON EMISSION FROM
FERROELECTRICS**

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Abstract

The efficiency of laser-induced electron emission from ferroelectric (FE) cathodes has been measured by illumination with a Ti:sapphire laser at a pulse length of 140 fs and is compared to earlier measurements at 40 ps and 5 ns pulse lengths. Two modes of emission are observed: (i) a "normal steady-state" emission in the presence of a constant extraction field and (ii) a transient mode after a polarization switching of the FE cathode by a high-voltage pulse. The latter mode leads to "self-emission" in high electron currents with kinetic electron energies of several kilovolts. The physical interpretation and the potential applications of laser-induced electron emission from FE cathodes are briefly pointed out.

1 Introduction

Laser-induced electron emission from ferroelectric (FE) material has been under study for several years [1]–[7]. The mechanisms of this type of emission differ strongly from conventional photoemission. The cathode materials used in the present and previous investigations are not fully ferroelectric at room temperature and in some cases not ferroelectric at all. FE microdomains can be formed under the influence of external (e.g. during polarization switching) or internal (e.g. space-charge field generation by pre-polarization of the photocathode) electric fields [8]. Two modes of laser-induced emission have been observed under different conditions of irradiation and extraction field application: a "normal steady-state" emission under the action of a constant extraction field and under a regular laser illumination, and a transient mode occurring after an interleaved polariza-

tion switching of the FE cathode with short high-voltage pulses [9]–[11]. The latter mode of laser-induced emission leads to a significant enhancement of the “normal” emission with extraction field [2] or to the “self-emission” of high electron currents with kinetic electron energies of several keV in the absence of an external extraction field [3]. Earlier investigations have shown an increase of emission efficiency by decreasing the laser pulse length from the nanosecond to the picosecond domain [7]. The aim of this work is to investigate the efficiency of normal and transient electron emission from FE photocathodes under illumination with femtosecond pulses and to compare their behaviour with that of metallic photocathodes.

2 Experimental set-up

The FE photocathodes were mainly made from zirconium-rich lead-lanthanum zirconium-titanate ceramics (for example, PLZT $x/y/1 - y$, meaning $[\text{Pb}_{1-x}, \text{La}_x][\text{Zr}_y, \text{Ti}_{1-y}]\text{O}_3$, where x and y are the fractional compositions of La and Zr). Single crystals of this type are optically transparent between 370 nm and 10 μm wavelength and absorb strongly outside this range. Laser irradiation was performed with a femtosecond Ti:sapphire laser including chirped-pulse regenerative amplification. The laser source (model CPA-1, CLARC, USA) provides pulses of 140 fs at 780 nm with a repetition rate of 1 kHz. The maximal available pulse energy was 0.6 mJ. The second harmonic of 390 nm could be generated in an LBO non-linear crystal with pulse energies up to 50 μJ . An electro-optic pulse generator was installed in order to select single pulses or any pulse frequency up to 1 kHz. The pulse energy could be reduced by optical attenuators and measured by a power meter or by pyroelectric joulemeters. Fast photodiodes and an intensity second-harmonic autocorrelator were used for the diagnostics of the laser pulses. The beam diameter at the output of the laser was 3.2 mm. It could be reduced by means of a beam telescope and a focusing lens to 0.5 mm. In all experiments the FE cathodes were installed in the same vacuum tank which had been used earlier in experiments with nanosecond and picosecond lasers. The angle of incidence of the laser beam on the cathode surface could be varied between 0° and 50° . The vacuum in which most tests were carried out was in the range from 10^{-6} to 10^{-5} mbar. Extraction voltages up to 35 kV could be applied between the sample support and the anode corresponding to d.c. extraction fields from 0 to 3.5 MV/m. The electron charge emitted from the cathode was measured directly when experiments with extraction voltage applied to the anode were performed. In self-emission experiments without extraction voltage the electrons collected at the anode were measured via an HV decoupling capacitor and a matched integrator to the input of an oscilloscope of 1 GHz bandwidth. The FE cathode support in the vacuum tank was adapted to FE polarization switching interleaved with laser irradiation. Several of the FE photocathodes studied in the fs tests have been investigated before with Nd:YAG laser systems at nanosecond and picosecond pulse lengths, which have been described earlier [4, 7].

3 Experimental results

The emission efficiency of FE photocathodes defined as $\eta = \text{emitted charge } Q / \text{incident laser energy } E$ (nC/mJ) is strongly increasing when going from 5 ns to 140 ps laser pulse length. Several photocathodes made from various polycrystalline ferroelectric materials have been investigated at a laser pulse length of 140 fs, at two wavelengths λ of 390 and 780 nm and at an extraction potential of up to 30 kV. Figure 1 shows the emission efficiency η of a PLZT 1/92/8 cathode as a function of power density at $\lambda = 780$ nm

under the extraction potential of 30 kV. The efficiency initially increases with more than the third power of laser power density, but then is limited by space charge. Figure 2(a) and 2(b) shows the experimental data, obtained in three different laboratories, with three different lasers, and at three different times, with one and the same FE photocathode, compressed into single diagrams. Figure 2(a) covers 7 orders of magnitude in incident laser power density W and shows the efficiency η in nC/mJ of this cathode, PZT 95/5 doped with 2% Cr_2O_3 , as a function of W in the whole range of pulse lengths and wavelengths covered by the three experimental series. The rate of efficiency enhancement with power density becomes higher for longer wavelengths, particularly at shorter laser pulse length. The slight offsets of the efficiency plots at λ of 266, 355 and 532 nm are attributed to the quite different experimental conditions rather than to a physical reason. The authors consider the branches as modestly well overlapping, hence, outside the space-charge region the efficiency depends on power density in the same way regardless of whether power density is achieved by focusing of the laser beam or by shortening of the pulse length. All efficiency curves valid at 30 kV extraction voltage reach a maximum with increasing power density. This limitation of efficiency can be attributed to space charge as shown in Fig. 2(b). Together with the measured data, the calculated space-charge limits are plotted as a function of power density with the laser pulse length as a parameter, taking into account that the transition time of the charge emitted from the photocathode surface is longer than the laser pulse. In our experiment this is valid up to a laser pulse length of 240 ps according to Ref. [12]. The maximum η that can be reached under space-charge conditions can be represented in a logarithmic scale by

$$\log \eta = \log k(V_{\text{ex}}) - \log t_p - \log W, \quad (1)$$

where $k(V_{\text{ex}})$ is a function depending on the external extraction potential V_{ex} and t_p the laser pulse length. Hence, contrary to the efficiency in the non-space-charge-dominated region, the calculated and experimentally observed space-charge limits increase not only for higher extraction voltage (see Fig. 3), but also with decreasing laser pulse length [see Fig. 2(a) and 2(b)]. Figure 4 shows a plot of emitted charge density as a function of laser pulse energy density for a PLZT 1/92/8 cathode and for a thoriated (2%) tungsten cathode of 2 mm thickness at 780 nm wavelength. The emitted charge of the thoriated tungsten cathode depends quadratically on the laser pulse energy indicating correctly the effect of two-photon excitation.

The emission efficiency in saturation of certain classical photocathodes is a material constant (quantum efficiency) in a limited range of wavelength, laser power density, or pulse length. This is not the case for the laser-induced emission efficiency of FE photocathodes. When comparing the results obtained with the three previously mentioned laser systems, several methods are recognized which allow the increase of the emission efficiency. Increasing the incident laser power density by decreasing pulse length and/or laser beam cross-section has the strongest influence at all wavelengths and laser beam incidence angles. Therefore with femtosecond laser pulses at 780 nm in the 10–100 GW/cm² range, the same efficiencies were reached as with UV pulses at low power density [Fig. 2(a)]. The rate of efficiency increase is more strongly marked at longer wavelengths than in the UV. In the whole range of wavelength and power density, covered by Fig. 2(a), and for the pulse lengths of 40 ps and 140 fs, the emission efficiency was finally limited by space charge (modest extraction field of only 3.5 MV/m) and available laser power, but not by destruction of the FE cathodes. Like the space-charge limit, the destruction limits also

Figure 1: Emission efficiency η of a PLZT 1/92/8 photocathode as a function of laser-power density striking the surface at normal incidence, at a laser pulse length of 140 fs, at a wavelength of $\lambda = 780$ nm and under an extraction voltage of 30 kV.

Figure 2: (a) Emission efficiency of a PZT 95/5 photocathode doped with 2% Cr_2O_3 for different laser wavelengths and pulse lengths as a function of laser power density at normal incidence and at 30 kV extraction voltage.

(b) Measured emission efficiency of the same cathode as a function of laser pulse power at three laser pulse lengths, 5 ns, 40 ps and 140 fs, and a 30 kV extraction voltage together with the calculated space-charge limits for each laser pulse length.

Figure 3: Measured emission efficiency of the PZT 95/5 photocathode with 2% Cr_2O_3 as a function of laser pulse power density for different extraction voltages at 140 fs laser pulse duration and at 780 nm together with the calculated space-charge limits for each voltage level.

Figure 4: Emitted charge density of a thoriated tungsten photocathode and a PLZT 1/92/8 cathode as a function of laser energy density at 30 kV extraction voltage and at 780 nm wavelength.

Figure 5: Self-emitted electron charge density from a PLZT 1/92/8 photocathode after polarization switching as a function of the number of laser pulses applied to the cathode at 780 nm, 140 fs, and zero extraction voltage.

decrease with shorter wavelength. The highest efficiencies η of normal steady-state emission observed with the femtosecond laser system were limited by space charge to about 1 nC/mJ. At higher extraction fields, more than 10 nC/mJ could have been reached before the destruction limit was met [Fig. 2(a)]. The maximum power density level reached in the femtosecond experiments was 1.7 TW/cm² on a PLZT 2/94.5/5.5 photocathode. After 15 minutes of irradiation with 1 kHz laser repetition rate, no visible damage could be observed on the surface area hit by the laser beam.

Enhanced (compared to “steady-state”) emission efficiency of more than 1 nC per mJ was observed with a PLZT 2/95/5 photocathode, which was newly inserted into the vacuum tank, and which had been used in previous experiments in the nanosecond and picosecond pulse length range. This enhancement, which was also observed with other FE cathodes, was of a transient nature. It decayed after several hundred laser pulses to the normal emission level and could not be reactivated since the switching capabilities of all FE cathodes were weak. It seems to be the same emission-enhancement process that was reproducibly observed formerly [2] as an order-of-magnitude effect with strongly offline-pre-polarized and well-switchable PLZT cathodes. The FE polarization switching was limited by the FE grid electrode structure, which was adapted to “normal, steady-state emission”, but not to HV-pulse excited emission. However, the polarization switching worked efficiently enough to reproduce laser-induced self-emission of 0.24 nC/cm² from a PLZT 1/92/8 cathode with 140 fs laser pulse length at 780 nm wavelength. Figure 5 shows the decay of emitted charge density with the laser-pulse number for zero extraction voltage after a weak polarization switching. Alternate switching and laser pulse irradiation maintained the emitted charge amplitude at a maximum level of 0.4 nC/cm². Self-emission was also observed at 390 nm wavelength and with other FE materials.

4 Discussion

The self-emission and enhancement effects are only observed after FE polarization switching as reported in Refs. [2] and [7]. Polarization switching includes not only P_s reversal, but also the creation or annihilation of FE domains by HV pulses. Transient amplified emission of charge densities, which can be achieved under normal (non-switching) conditions only at UV wavelengths, has again been observed in the visible and the IR region. During laser-induced emission experiments with FE photocathodes, it was also recognized that the application of a d.c. extraction field eliminates the polarization switching capability of the FE cathode in the region of the laser spot. The transient enhancement, which grows with increasing wavelength, is jeopardized under repetitive laser illumination accompanied by strong d.c. extraction potentials ([2] and [7]).

Whereas normal steady-state laser-induced emission is based not only on ferroelectric properties, self-emission is totally determined by the FE characteristics of the cathode. The distribution of kinetic energies of the emitted electrons had been roughly measured on a PLZT 2/94.5/5.5 cathode reported in Ref. [3]. The amount of average polarization ΔP_s switched in the photocathode can be expressed by

$$\Delta P_s = \frac{\delta^3 P''}{h F_s} \int_0^{F_s} \int_0^h m' n' dF dx, \quad (2)$$

where P'' is the average polarization of a single FE domain, F_s and h are laser spot area and cathode thickness, δ the FE domain size (typically $< 0.1 \mu\text{m}$), $n' = dn/dx$ the perpendicular and $m' = dm/df$ the transversal distributions of the switchable FE domains. The average kinetic energy is given to the electrons by the potential raised on the photocathode surface after the polarization change ΔP_s induced by switching with the laser pulse energy. The average potential V_{kin} can be described approximately by

$$V_{\text{kin}} = \Delta P_s a / \epsilon \quad (3)$$

where ϵ is the dielectric constant and $a = f(\lambda, T_{\text{FE}})$ is the average depth, down to which FE domains are switched by light with wavelength λ in an FE photocathode of optical transmission T_{FE} . Equation (3) shows that FE switching by laser may happen in a thin surface layer with a high ΔP_s , as is the case with UV light or with a strongly absorbing material; or at longer wavelengths with a lower ΔP_s and a larger depth a with a more transparent material; or at visible or IR wavelengths. The decay of self-emission with repetitive laser irradiation seems to be due to the different magnitudes of the polarization switching and of the charge compensation processes after an electric-field or after a laser excitation. In contrast with laser switching, HV switching acts on a major part of the cathode volume, and the subsequent surface charge compensation is a much more violent effect than after laser switching. Whereas the high surface field after HV switching leads to a rapid charge compensation via the emitting surface, laser-induced polarization changes can also be gradually compensated with internal space charge (internal bulk electrons and holes). The compensating electrons bound loosely on the surface can be emitted more easily from the FE cathode surface than electrons bound deeper in the matrix of the sample.

5 Conclusion

In conclusion, the normal emission efficiency of all known cathode materials seems to increase strongly with growing power density. The use of ultrashort femtosecond laser

pulses is favourable for photocathodes, especially for ferroelectric ones. The “basic” laser-induced emission efficiency of FE photocathodes surpasses that of metallic photocathodes by at least one order of magnitude. The difference depends on wavelength and is more important in the visible and in the IR regions than in the UV region. The maximum peak-power density level reached in the femtosecond experiments with FE photocathodes was 1.7 TW/cm^2 . After 15 minutes of irradiation of a PLZT 2/94.5/5.5 cathode with 1 kHz repetition rate, no visible damage could be observed on the surface area hit by the laser beam. With respect to reliability and lifetime, FE photocathodes are superior to most metallic photocathodes, and they can work well in a gas or plasma atmosphere. As in the case of electric-field excited self-emission from ferroelectric cathodes, laser-induced FE self-emission also allows the construction of electron guns featuring the spatial separation of electron generation and of their acceleration. Ferroelectric photocathodes seem to be increasingly useful for many types of high-power electron guns, including applications in free-electron lasers and future high-power and high-repetition-rate linear colliders.

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