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Photocathodes: the state of the art and some news

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Abstract

The present cesium telluride cathodes have the capability to provide, for months, trains of picosecond pulses carrying a charge equal/higher than 10 nanocoulomb, for a total charge higher than the microcoulomb. However, they are very delicate. A possible way to increase their robustness is by covering them with a protective film of nanostructured carbon. Ferroelectric ceramics, as possible new robust photocathodes, showed an interesting level of emission with 532 nm–25 ps laser pulses.

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1. Introduction

High-luminosity accelerators and short-wavelength free electron lasers require RF-guns with robust photocathodes of high-yield, low emittance and negligible dark current [1]. The today cesium telluride Cs_2Te photocathodes operated for 3–6 months with a QE higher than 7% and for about two years with a QE higher than 2%, in an RF-gun with a vacuum of ~ 0.5 nTorr, an accelerating field $E_{acc} \sim 100$ MV/m. They were shined with the 262 nm (4.7 eV) light and delivered a charge $C = 400$–640 nC/train with 10 nC/bunch [2–4]. These cathodes do not survive to even a very short exposure to low–medium vacuum ($p \sim 10^{-7}$ mbar). When the transferring a cathode from the preparation chamber to the RF-gun cannot be done under vacuum, the more robust cesium iodide with 2 nm germanium coating is used. The QE of these cathodes when fresh is $\sim 0.5\%$ (shined with the 262 nm), but after a year of operation it is still at the level of 0.3% [2]. Potassium, rubidium and rubidium–cesium telluride, K_2Te, Rb_2Te, RbCsTe, cathodes operate routinely in RF-gun with high efficiency. Furthermore, they can be rejuvenated after air exposure, recovering a QE of 0.5% [2,5]. All the above cathodes have shown a picosecond response time and no bunch lengthening.

In search for a robust photocathode, a diamond-based photoemitter has been tested [6], a ferroelectric photoemitter is under investigation [7] and a protective film of nanostructured carbon onto the alkali photocathodes has been proposed [8]. A boron-doped polycrystalline diamond sample, grown on a molybdenum substrate, operated in a vacuum environment as high as 10^{-4} Torr, showed a quantum efficiency of

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3×10^{-4} at $\lambda = 193$ nm (6.4 eV). Ferroelectric ceramics seem interesting because the polarization induces on one surface 100 $\mu C/cm^2$ of compensating space charge packed in a film of a thickness of the order of 50 nm and on the opposite surface the polarization induces a downward band bending which reduces considerably the work function. Nanostructured carbon film promises to have a good transmittance of both the incident light and the substrate emitted electrons, it is an inert material and, nevertheless, it bounds solidly to the substrate [9].

The generalized Fowler–DuBrige–Bechtel et al. theory successfully explains the photoemission at the different regimes up to a certain incident light intensity [10]: the one-photon with dependence from temperature and/or from the photon frequency (energy), the multi-photon, the thermally assisted multi-photon and the two-temperature multi-photon regime.

The emission process encompasses the microsystem–photon(s) interaction (with photons absorption and the consequent energetic electron separation), the migration of the separated electron toward the surface and the final escape from the material. The basic assumption of the model is that the electron emission current density J is the sum of the partial currents relative to the n-photon emissions

$$J = \sum_{n=0}^{\infty} J_n = \sum_{n=0}^{\infty} \sigma_n I^n.$$

Here, J_0 is interpreted as thermionic emission, J_1, J_2, etc. as one-photon, two-photon, etc. photoemission, I is the laser intensity; the generalized n-photon ionization cross section σ_n in the nth component lumps the dependence on temperature (governed by the heat conduction equation [10]), reflectivity, transition probability and escape probability.

It has been experimentally observed that a net enhancement of the photoelectric efficiency occurs when a metal is illuminated with a very intense ultra short laser pulse [11]. The slopes of the emitted charge versus laser intensity are higher than that predicted by the n-photon regime. In order to explain this behavior, it has been suggested that the Auger effect, that is the energy transfer between two colliding electrons with the dropping of one of the two into a lower depleted level, should be added to the photon absorption [12]. When the exciting light pulse is equal or shorter than the electron–phonon relaxation time τ_{ep} and the photon density is so high that the density of the hot electron cloud is higher than $10^{20}/cm^3$, the probability of an Auger collision becomes higher than the probability of a multi-photon absorption. In fact, the density of the hot electrons becomes higher than that of the incident photons because they accumulate within the narrow absorbing surface sheet [8].

2. The work on photocathodes

2.1. Single photon photocathodes

In searching for a photoemitter of high current density (hundreds–thousand amperes/cm^2), high charge in a multibunch trains (500–1000 nC/train with bunchlets of some picoseconds), fast response ($\leq$ ps) and robustness, people worked on metallic and/or semiconductor photoemitters. The former have shown good characteristics (the high peak current deserves a particular mention) except for the yield: this resulted at best 10^{-4} electrons per incident UV photon (the escape depth is few Å and the surface absorption is poor). The yield is too low for the required charge in the main projects. In addition, it is reported that the emittance could be relatively high (due to e–e scattering) [13].

The alkali antimonide, telluride and halide photoemitters, together with gallium arsenide, have been extensively studied and technologically developed for their capability to deliver the required charge. The high QE is explained mainly by two facts: the high ratio between the gap energy E_g and the electron affinity E_a and the long escape depth (hundreds of Å) in conjunction with good light absorption within that layer.

The alkali antimonides Cs_3Sb and K_2CsSb were the first materials tested as photocathodes. Although these cathodes could operate in the visible range, they are dismissed because they require ultrahigh vacuum ($p \leq 10^{-10}$ mbar), are short

living and their performance depends on the "history" of the substrate.

Cesium iodide. This material is easy to prepare and robust (it resists exposure to air for a short time), but has a work function of $E_g = 6.3$ eV ($E_a =$ 0.1 eV). It was found that cesium iodide with 2 nm coating of germanium improves its quantum efficiency at 262 nm up to 0.5% and it resists exposure to air. The emission saturates at a certain level [1].

The cesium iodide cathode is prepared by depositing on an aluminum substrate 350 nm of CsI and then a coating of 2 nm of Ge. 15 h of conditioning (with high electric fields and laser pulses) is required.

Cesium telluride. This photoemissive material is considered at present time the best among all for its characteristics of robustness, high QE, stability of emission for months, capability to operate with the fourth harmonic of the Nd:YAG laser (which is bad but not impossible) and, finally, the capability to rejuvenate after deterioration (for exposure to 10^{-4} mbar for hours) [14].

The Cs_2Te is prepared by depositing about 10 nm of Te and in succession on a film of Cs (so as to maximize the photocurrent onto a molybdenum substrate kept at 120°C. The final Cs_2Te film thickness results in about 30 nm. The vacuum in the preparation chamber is maintained better than 10^{-10} mbar during the evaporation process [14]. Each lab has developed its own receipe for cathode preparation.

Potassium, rubidium and rubidium-cesium telluride: K_2Te, Rb_2Te, RbCsTe. K_2Te photocathode is less efficient than others, but more robust. The other two are comparable with Cs_2Te, but their rejuvenation after air exposition is better [2,5].

The laser systems (mostly Nd:YAG and Nd:YLF systems) used to illuminate photocathodes [1] are reliable but they still have some problems: the stability of alignment is difficult, the characteristics of the components (Pockels cells, Faraday Rotators, grating and mirrors) are not stable. The mechanical design and the quality of the mounts maybe need further improvement. The operational high-power density (10 GW/cm^2) in connection with the unavoidable ambient dust are responsible for component damage. A time-to-time substitution of components must be assumed. An every day check and resetting of the laser is unavoidable. New ideas towards a reduction of the energy density would be welcome. In this context of stability and reliability of the laser system, it must be noticed that a very complex laser system lives near or inside a gigantic equipment ambient. The laser people under the pressure for a stable system suggest to move towards the green light. The first harmonic, they say, is much better controllable.

2.2. The multiphoton photoemission and the relative thermally assisted regime

The photoemission via two-photon interaction is worth considering because it allows the use of metallic-ferroelectric cathodes and the use of relatively soft photons, in the green–red range of the electromagnetic spectrum. In that range, the energy/power per pulse provided by lasers is a factor three–ten higher than the UV light, depending on what harmonic is compared to, in addition, the reflected light (from the cathode) affects the RF-cell wall less.

The strong drawback is the very low yield, QE $= 10^{-6}$–10^{-9} depending on the material and the incident intensity. Considering the best results, the robustness of metallic photocathodes and considering that the reduction of the laser power in passing from the second to the fourth harmonic is higher than one order of magnitude, it can be concluded that a material with a QE of 10^{-4}–10^{-5} operating in two-photon regime becomes interesting. Anyway, that kind of material operating at the second harmonic, is a good candidate as cathode in cases where the upper limits of charge and multipulse timing are not required. This argument is stronger when the fifth harmonic is required. We can guess that the emittance is the drawback of this regime (no measurement is done yet). A measurement of thermal emittance is programmed at LASA-Milano Lab.

3. New ideas

3.1. A protective film on alkali photoemitters

In pursuing the purpose of strengthening the present good alkali photoemitters, we propose to

cover the photosensitive material with a protective film of nanostructured carbon [8]. The choice of nanostructured carbon film comes from the lucky combination of chemical inertness with a good transmissivity of the light (at least at 20 nm thickness) and a good conductivity (which comes from the dominant sp2 (graphite-like) character [15,16]). Moreover, that carbon film turns out to be strongly bound to the substrate [9].

We started to check the effect of that kind of film on Cu and Au substrates using the green 2.33 eV light ($\lambda = 532$ nm). Since the emission yield depends on the film thickness, we made measurements with different film thicknesses: 20, 70, 170, 200 nm. We have checked, with a SEM microscope, that the 20 nm film covers completely the substrate. In Fig. 1, the emitted charge as function of laser pulse intensity for a gold disk coated with 20 nm film and for a plain gold disk is shown. The two emission curves almost overlap. The slope is around two at lower points, then it increases around 4. This indicates that the emission, at a fluence higher than 100 MW/cm^2 for 25 ps, is of the Auger type [8].

The emission curves obtained with plain and covered copper gave the same result. Instead, the emission of Cu covered with 70 and 170 nm film differs. Their emissions, hence, did not depend on the substrate.

The adhesion of the films onto the substrate remained stable after many thousands of shots as long as the intensity of the light was below 1 mJ/10 mm^2 at the 25 ps long pulses and with a repetition rate of 10 Hz.

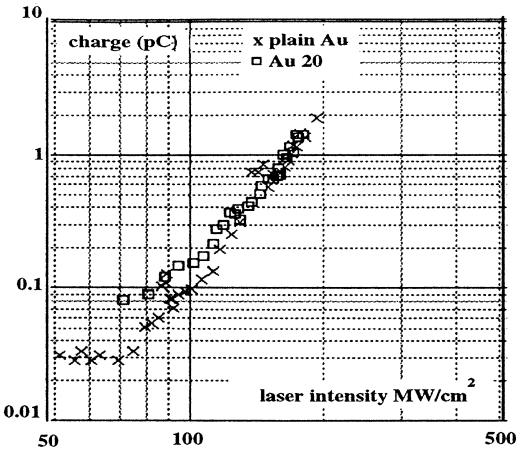

Fig. 1. Charge emitted versus energy of the laser pulse obtained with a cathode of plain gold (◇) and gold coated with a 20 nm film of nanostructured carbon (+). The slope in log–log diagram results in 4.3.

3.2. Ferroelectric polarized ceramics as photoemitters

Ceramic is a material resulting from the sintering process of oxides. Its characteristics can be determined: (a) changing the percentage of the different components, (b) changing the temperature and time of the sintering process, (c) by prepoling the disk (which means making n-or p-type the surfaces), (d) by the level of polarization (which means bending upward or downward the energy bands at the surfaces) and finally by (d) the temperature (which means changing the resistivity, the polarization and the response velocity to excitation) [17].

Ceramics can withstand over 30 mJ/cm^2 of 2.33 eV–30 ps laser pulse, which is at least a factor three more than a metallic photocathode. A positive polarization accomplishes the downward band bending, hence shifts down the vacuum level, so, in turn, reduces the work function. The idea is to reduce the energy gap/affinity ratio E_g/E_a to a convenient value by finding a lucky composition, a proper sintering and by exploiting the prepoling and the polarization.

The experiments has been carried out, so far, with lead zirconate titanate doped with lanthanum and with chromium [18]. We get 1 nC/10 mm^2 with 3 mJ pulse energy at $\lambda = 532$ nm and 100 pC/2 mm^2 with 0.3 mJ at $\lambda = 355$ nm, see Refs. [7,18]. In the first experiment the emission was clearly space charge limited. The extrapolation would have given a charge of 2 nC and a quantum efficiency of 10^{-6}. The two results were comparable. Those emissions were obtained with unprepoled samples and the polarization was induced by the accelerating field only. A similar result, see Fig. 2, was obtained in completely different experimental conditions: the cathode disk had the front surface electroded with a metallic grating of 200 μm wide stripes inter-distanced by the same

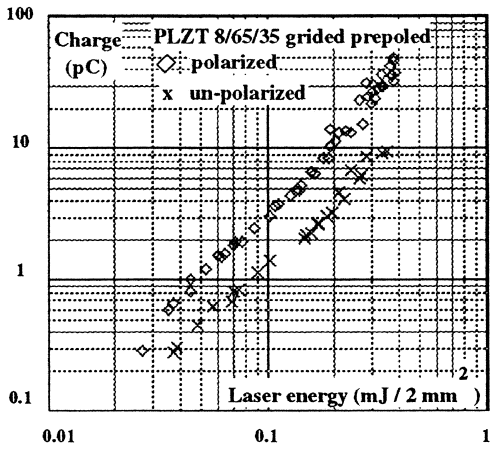

Fig. 2. Emitted charge versus laser energy for PLZT 8/65/35 illuminated with violet light in log–log plot. The slope of the line is 2, for $Q \alpha I^2$.

width, it was prepoled and finally it was polarized with a bias of 1 kV/mm applied between the front and the back electrodes (with the positive side on the front electrode). The polarization orientation was the same as induced by the accelerating field.

The results are promising because they show that the emission is controllable by external parameters, the light can be in the optical range and the quantum efficiency is considerably higher than in metallic cathodes.

Follow up experiments will focus on photoemission from ceramics as a function of temperature, film deposition on a substrate, substrate ceramic contact potential, different materials.

4. Conclusion

The Cs_2Te, K_2Te and KCsTe satisfy the present requirements. They are only difficult to manage. The issues about these photocathodes are: (1) keeping the emitting area homogeneous; (2) lowering the dark current to 50 nA at the operating RF; (3) measuring the thermal emittance; (4) and measuring the promptness of the photoemission process for picosecond bunches.

The protective film of nanostructured carbon must be checked on an alkali cathode. If it will behave as a neutral protective sheet, the alkali antimonides, as Cs_3Sb, could be reconsidered so as to move towards the green part of the light spectrum.

About ceramics, the research is at the beginning. The first findings are interesting and promising.

The Auger Effect explains the behavior at photocathodes at high laser intensity.

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