

## EXPERIMENTAL BEHAVIOUR OF CESIUM-ANTIMONY PHOTOCATHODES UNDER LASER IRRADIATION

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### Abstract

The behaviour of  $Cs_3Sb$  photocathodes was observed when illuminated by a CW  $Ar^+$  laser. The density of cesium on the surface and the thermal gradient induced by the laser are clearly bound. The maximum of the current intensity corresponding to the limit of linear photoelectric response of such photocathodes was estimated to  $< 100 \text{ mA/cm}^2$  at 500 nm, in complete disagreement with previous optimistic data.

### 1 - Introduction

Photocathodes are traditionally used in radiation detectors and imaging tubes for the detection of low-light levels with photocurrent densities below  $1 \mu\text{A/cm}^2$ . During the last decade, photocathodes have emerged as a means of generating high-current-density, bunched-electron beams with low-emittance, required for efficient operation of rf-Linac-driven free-electron lasers [1-3]. Laser-driven photoemitters appear to meet these specifications [4]. Various metals [5] and semi-conducting materials have been tested, these last presenting for visible light an higher photoelectric yield. A comparison between the optical performances of typical photoemissive materials used in photo-injectors was reported in ref. [6]. Among semi-conducting photocathodes, cesium-antimony received a special attention [7-10], because efficiency up to 20 % can be observed for  $\sim 350 \text{ nm}$  radiation. This very high efficiency is attributed to their cubic structure [11]. Such a structure is supposed stable when the temperature is less than  $650^\circ\text{C}$ , but the melting temperature of cesium is only  $28^\circ\text{C}$ . The rather tricky methods of preparation are described in ref. [11]. However, it has been previously observed that high levels of radiative exposure of such a material results in fatigue of the photocathodes, so that they can only be operated for a short period of time. In order to estimate their effective performance and their limit of linear behaviour, we have performed a series of experiments. Although we used a CW laser source, these results can be related to the action of bunches with a very high repetition laser-pulse-rate [12].

### 2 - Apparatus Design

Figure 1 shows a schematic diagram of our testing apparatus. The major components are : a vacuum diode containing the photocathode, the radiation source, the optical beam transport components and an ammeter for the photocurrent-intensity measurement. We used three different professional photodiodes referenced as AVHC 40 (DARIO), AVHC 41 (RTC) and F 4000 (RTC) with typical responses S4 and S5. The cesium antimony was deposited as polycrystalline thin shells on a plane metallic substrate and enclosed in a clean, free of oxygen, vacuum electronic tube. At room temperature Cs vapor is developed in the diode with a pressure  $\sim 10^{-6}$  torr. For each one we verified that they had photoelectric performances very close to the best data

reported in the literature, proving the initial high quality of the active surfaces. The plane anode constituted by a mesh of very thin wires is parallel to the cathode just behind a plane glass window of 0.5 mm thickness.

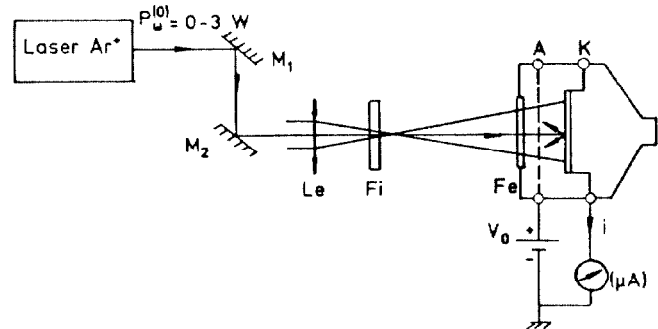


Figure 1 - Diagram of the testing apparatus

These diodes support up to 5 kV but we limited the applied voltage to 3 kV because the saturation working regime is effective from 300 V.

The radiation source was a CW  $Ar^+$  laser with  $P_{\omega}^{(0)} = 5 \text{ W}$  the maximum output power at  $500 \pm 14 \text{ nm}$ . However, the incident laser power was limited to 3 W to prevent a too much rapid destruction of photocathodes. Because the spot-diameter of the laser-beam changes with the emitted power, the illuminated area on the photocathodes varied from 2 to 6  $\text{cm}^2$ . The same laser was used to measure the photoelectric sensitivity of  $Cs_3Sb$ , but in this case, we reduced the laser intensity on the photocathodes by a factor  $10^4$ , with neutral density filters. All measurements were done with the laser beam arriving at right angle on the photocathodes. The photocurrent intensity was measured with a pico-ammeter Keithley 414 S ( $10^{-10}$  to 1 A) directly connected to the cathode electrical circuitry.

### 3 - Experimental data

For each vacuum diode, effective photoelectric sensitivity of the active layer was preliminary performed. The current intensity  $i_s$  was detected for an applied voltage  $V_0$  varying between 300 V and 3 kV. As shown on Figure 2,  $i_s$  appeared to vary linearly with  $V_0^{1/2}$  and because the very small slope of the curves, we can assert that the diodes were working in Schottky regime.

The values of the sensibilities given below :  $S_{\omega}^{(0)}$ , correspond to an applied voltage  $V_0 = 0$ .

The absorbed laser intensity  $I_{\omega}^a$  can be calculated from the simplified expression

$$I_{\omega}^a \approx 0.95 \frac{T_F \times P_{\omega}^{(0)}}{d^2} \quad (1)$$

where  $T_F \sim 10^{-1}-10^{-5}$  is the transmittivity of the neutral filter and  $d$  the beam diameter on the cathode.

So, we observe the change of  $S_{\omega}^{(0)}$  with  $I_{\omega}^a$  even at very low laser intensity. We think that is due to the variation of the illuminated photocathode area  $S$ . The lowering of  $S_{\omega}^{(0)}$  when  $I_{\omega}^a$  is increased corresponds to the inhomogeneity of the active layer surface. This point was observed with the three tested cathodes and constitutes a further difficulty for a practical application in photo-injectors.

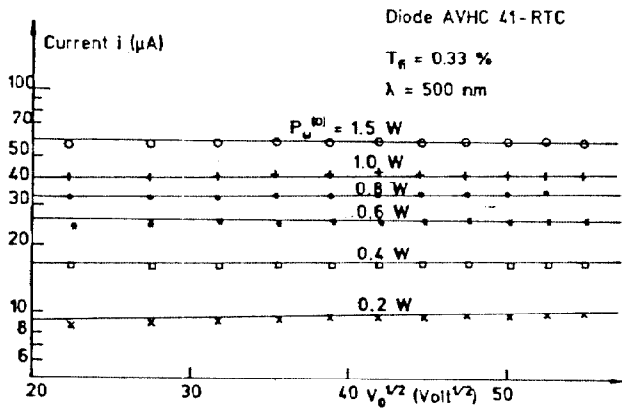


Figure 2 - Current-Voltage characteristics of AVHC 41 RTC diode

Now we consider the photocurrent variations versus the time and the absorbed laser intensity. Typical responses are reported on Figure 3, the characteristics of target illumination being given Table I.

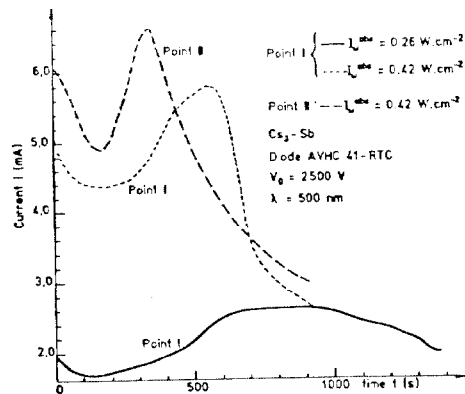


Figure 3 - Current intensity versus time of AVHC 41 RTC diode for two different points of the photocathode surface and various absorbed laser intensity

The current  $i_s$  was measured every 10 seconds while  $P_{\omega}^{(0)}$  was kept constant. Figure 3 shows that for a relatively small value of  $I_{\omega}^a < 0.2 \text{ W/cm}^2$ ,  $i_s$  varies monotonously, while for higher laser intensity,  $i_s$  presents a large modulation. We think that it is typical of the following situation : during the first minutes of the irradiation, the growth of the surface

$P_{\omega}^{(0)}$ (W)	$d^{(*)}$ (mm)	$S$ (cm <sup>2</sup> )	$I_{\omega}^a$ ( (W/cm <sup>2</sup> )
1.0	20	3.14	0.24
1.5	21	3.46	0.33
2.0	23	4.15	0.36
2.5	25	4.9	0.38
3.0	26	5.3	0.42

(\*) The relative precision on  $d$  is 5 %, so that the precision on  $I_{\omega}^a$  is  $\approx 10 \%$  .

Table I - Values of  $P_{\omega}^{(0)}$  and  $I_{\omega}^a$  related to the Figure 3

temperature induced by the laser beam leads to a progressive cesium desorption at the surface.

The vapor pressure in the tube increases up to  $\sim 10^{-4}$  torr [11]. The principal consequence of the Cs degeneration is the net reduction of the active surface sensibility. Some four minutes later, the temperature gradient in the bulk of active material is then sufficient to cause the migration of Cs up to the surface, and to reform the cesium surface monolayer. As a consequence, the sensibility increases again. But correlatively the Cs at the surface being rapidly desorbed, because of the high temperature induced by the laser beam, when all the atoms of Cs present in the inner shells, have been desorbed, we observe the definitive reduction of the photoelectric sensibility, so that less than 15 mn after the beginning of the photocathode illumination, the sensibility is  $< 1 \text{ mA/W}$ .

While at very low laser flux,  $S_{\omega}^{(0)}$  was  $\sim 18.5 \text{ mA/W}$ , the maximum of sensibility corresponding to the peak of current on Figure 3 is only  $\sim 2.9 \text{ mA/W}$ . The photocathode with AVHC 40 DARIO tube shows an higher resistance which can be certainly connected to the know-how for its realization. But, the same evolution was observed, for a comparable illumination duration to that of the former tube. To be sure that the reduction of sensibility depends effectively on the heating of the cathode, we measured the duration needed to find again a given photoelectric sensibility after absorption of a calibrated laser fluences. So, we realized four successive illuminations of the AVHC 40 photocathode during 1, 2, 3 and 4 minutes, corresponding to absorption of 57, 114, 172 and 229 J/cm<sup>2</sup> respectively. The photoelectric sensibility could be measured only during the periods between two consecutive irradiations. During each irradiation,  $S_{\omega}^{(0)}$  is supposed to varylinearly. Figure 4 illustrates our observations. We note a rapid reduction by a factor  $\sim 2$  of the current intensity  $i_s$  after the two first consecutive illuminations. Following each irradiation, we observe a small growth of  $i_s$ , which can be assigned to the partial reformation of the Cs-upper-monolayer. This is only possible because the presence of the low pressure Cs vapor in the diode. When  $i_s$  is again  $\sim 0.6 \mu\text{A}$ , 120 s after the end of the second irradiation, we proceed to the third illumination during 3 mn. Then, we observe a new reduction of  $i_s$  and  $S_{\omega}^{(0)}$ , but now the duration needed to partially reform the Cs-layer is in order to 390 s. After a fourth illumination during 4 mn, the duration needed to find again  $i_s \sim 0.6 \mu\text{A}$ , is about 1200 s. Similar behaviour was observed for the RTC F 4000 tube.

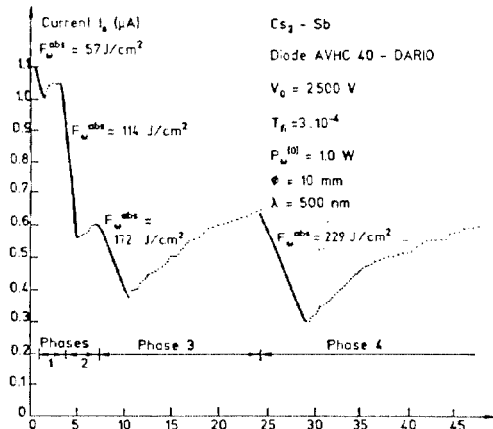


Figure 4 - Current intensity versus time of AVHC 40 DARIO diode for four laser fluences ( $F_w^a$ ) absorbed consecutively

All the experiments confirm the fragility of the Cs<sub>3</sub>Sb and our results are in good agreement with the previous data reported by Garbuny et al [13], on the change of sensibility with T from 10.4°C to 45°C.

To verify how  $S_\omega^{(0)}$  is effectively correlated to the thermal gradient of the active layer, we can make a relatively simple evaluation of the cooling duration between two successive illuminations of a material. We know that heating gradient of a surface is proportional to the heating duration t :

$$\Delta T = T - T_i \propto \sqrt{t} \tag{2}$$

while the natural cooling gradient is :

$$\Delta T = T' - T_M \propto \sqrt{t'} - \sqrt{t_M} - \sqrt{t' - t_M} \tag{3}$$

where  $T_i$  and  $T_M$  are the initial (room) and maximum temperatures of the surface, and  $t'$  is greater than  $t_M$  corresponding to  $T_M$ . Considering our heating conditions, the cooling of the surface to a given temperature, deduced from eqs (2) and (3) corresponds to 120, 300 and 600 s respectively. These values are to be compared with the experimental durations from Figure 5 : 120, 390 and 1200 s. These two sets of data show a comparable progression and are in relative agreement. The observed differences can be connected to the chemical reactions of the surface with its surrounding vapor, which are not taken into account in eqs. (2) and (3).

At last, the sensibility of these tubes a week after the end of these series of irradiations shows that  $S_\omega^{(0)}$  was definitively reduced for each one by a factor > 4, proving the irreversibility of the Cs<sub>3</sub>Sb chemical transformation at the surface.

#### 4 - Conclusion

Our experimental data are in complete disagreement with the previous one of Lee and al [9]. According to them, an uncooled Cs<sub>3</sub>Sb photocathode in a vacuum chamber would operate at 2.9 A/cm<sup>2</sup> and the cathode life would exceed 50 h. Their conclusions are wrong because they experimented

Cs<sub>3</sub>Sb photocathodes with a much lower power Ar<sup>+</sup> laser, typically 25 mW and they irradiated a surface much smaller than 1 cm<sup>2</sup>. Any extrapolation in this way, it not possible. The deposited laser energy in their experiments is much smaller than the quantities used in our work, so that the behaviours of Cs<sub>3</sub>Sb cathodes are completely different. Then, our first conclusion is that to preserve the photoemissive properties of Cs<sub>3</sub>Sb illuminated area > 1 cm<sup>2</sup>, the irradiation intensity must not exceed 100 mW/cm<sup>2</sup>. With a typical quantum efficiency  $\rho_Q \sim 5\%$  at 500 nm observed only for a low energy radiating source, the peak current is no more than 5 mA/cm<sup>2</sup>. It is always possible to produce an higher current density in increasing the incident laser intensity. But in this case,  $S_\omega^{(0)}$  will be reduced and for instance, with 1 W/(cm<sup>2</sup>) radiating density after some minutes,  $\rho_Q < 1\%$ , so that  $i_{max} \sim 10$  mA/cm<sup>2</sup>. The gain is only 2, while the absorbed laser energy was improved by 10. A surface illuminated by a high-frequency-laser-train, will be partially cooling between two consecutive laser pulses and the linear limit will be certainly higher than the one indicated above. However, pulse-laser-train induces mechanical stresses, so that the upper limit of the mean current density is closed to 100 mA/cm<sup>2</sup> for Cs<sub>3</sub>Sb.

#### References

- [1] D.T. Pierce et al., *Rev. Sci. Instrum.*, vol. 51, p. 478 1980.
- [2] C.K. Sinclair et al, *IEEE Trans. Nucl. Sci.* NS-28, p. 2649, 1981.
- [3] M. Yoshioka et al, *Proceedings of the 1984 Linear Acceleration Conference*, Seeheim, Germany, 1984, p. 469.
- [4] J.S. Fraser et al, in *Proceedings of Orsay Workshop*, June 30-July 3, 1987, S. Turner Ed., C.E.R.N., Rep. 87-11, Oct. 1987.
- [5] S.W. Downey et al, *Appl. Phys. Lett.*, vol. 49, p. L911, 1986.
- [6] J.P. Girardeau-Montaut et al, *J. Appl. Phys.*, vol. 65, p. 2889, 1989.
- [7] C.H. Lee, *J. Appl. Phys.*, vol. 54, p. 4578, 1983.
- [8] C.H. Lee, *Appl. Phys. Lett.*, vol. 44, p. 565 (1984).
- [9] C.H. Lee et al, *Rev. Sci. Instru.*, vol. 56, p. 560, 1985.
- [10] P.E. Oettinger, I. Bursuc, R.E. Sheffer and E. Pugh, *Appl. Phys. Lett.*, vol. 50, p. 1867 (1987).
- [11] A.H. SOMMER, *Photoemissive material*, Huntington, New York, R.E. Krieger Publ., 2nd edition, 1980.
- [12] C. Girardeau-Montaut et al, in *Laser surface treatment of metals*, C.N. Draper and P. Mazzoldi Ed., Boston, Martinus Nijhoff Publish., 1986, p. 235-248.
- [13] M. Gaburny et al, *J. Opt. Soc. Am.*, vol. 51, p. 261, 1961.