CATHODES FOR PHOTOEMISSION GUNS*

L. Cultrera#, CLASSE, Cornell University Ithaca NY 14853

Abstract

The last decade has seen a considerable interest in pursuit and realization of novel light sources such as Free Electron Lasers and Energy Recovery Linacs that promise to deliver unprecedented quality x-ray beams. The performance of these machines is strongly related to the brightness of the electron beam generating the x-rays. The brightness of the electron beam itself is mainly limited by the physical processes by which electrons are generated. For laser based photoemission sources this limit is ultimately related to the properties of photocathodes. In this paper an overview of the recent progress on photocathode development for photoemission electron sources is presented.

INTRODUCTION

The electron sources for advanced projects of fourth generation X-ray light sources, such as SASE-FELs [1], Energy Recovery Linacs [2], as well as future linear colliders [3], are mainly based on laser excited photocathodes placed in high electric field environment. Much recent progress has been achieved on the demonstration of accelerating schemes that allow generation of high charge density electron bunches with preserved beam emittance (e.g. see[4]). However, the ultimate electron beam brightness is strongly related to electron emission processes and the photocathode intrinsic properties [5]. Within this framework, research activities aimed at developing reliable electron photocathodes that have properties tailored towards specific accelerator applications have gained in importance over the last several years.

Laser photoemission offers several advantages for electron production such as is the ability to generate electron bunches of very short duration, down to few tens of fs [6], flexible time structure [7], and very low beam emittance [8].

Numerous materials have been used in photocathodes but is worthy to note that the experimental data (e.g. intrinsic or thermal emittance) are not complete and the agreement with the theory is not satisfactory in many cases [9].

PHOTOCATHODES PROPERTIES

The beam brightness is defined as the ratio between the beam current and the beam emittance (either in 2D, 4D or 6D phase space). Accordingly the way to increase the beam brightness is through the increase of the beam current, transverse emittance reduction, and/or response time. It is well known that the electron beam emittance forms as an interplay of different phenomena in electron sources. The photocathode's transverse velocity spread (or mean transverse energy, MTE) is one fundamental limit for high brightness electron generation.

Other relevant photocathode properties are the quantum efficiency (QE), - the number of electrons extracted per incident photon -, and the response time, i.e. the time it takes for the excited electron to escape from the photocathode surface. These properties determine the achievable bunch charge for a given laser intensity as well as the temporal shape of the emitted electron bunches. A prompt response time (< 1ps) is desirable for most photoinjector applications, while a much slower longer response time (10's of ps) is usually not acceptable unless a complex RF bunching and chopping system is being employed. Finally, a good photocathode should be uniform in its photoemission properties over the area of laser illumination and must be rugged enough to allow an acceptable operation time in the accelerator.

PHOTOCATHODES MATERIALS

Availability of reliable commercial laser sources with wavelengths ranging from IR to UV , with pulse durations ranging from few ns to few fs, and an average power up to few tens of watts opens the possibility for a number of material candidates [9], mainly metal- and semiconductor-based photocathodes. Furthermore, the semiconductor photocathode devices are further classified into positive and negative affinity families.

The electron photoemission theory has been actively pursued for both the metallic and semiconducting materials seeking to explain QE and MTE in terms of material properties and for a given laser wavelength [10-12]. Among parameters of relevance to the accelerators, it is worth to note the relationship between the QE and MTE with respect to the laser wavelength, the effective work function (defined as the surface work function lowered by the Schottky effect due to the presence of an external electric field), which is in principle applicable to any photocathode with negligible inelastic collisions during electron transport. A higher QE as well as an increased MTE is achievable from metallic photocathodes in very high electric fields, [10, 11] though the experimental results frequently disagree with the theoretical predictions indicating the path for further investigations.

Metallic Photocathodes

Metallic photocathodes are routinely used in many low duty factor photoinjectors. Their prompt time response makes them suitable for generating single or trains of extremely short electron bunches [13, 14]. Metals surfaces have been long supposed to be rather insensitive to contamination, thus, offering an advantage for preparation, handling, vacuum requirements, and

*This work is supported by NSF under Grant No. DMR-0807731 and DOE under Grant No. DE-SC0003965.

#lc572@cornell.edu

operational lifetime as opposed to the semiconductor photocathodes. On the other hand, their rather high work function and strong electron scattering processes result in poor QE and laser requirement to operate in the UV region. The typical electron yields for Cu and Mg, the most widely used metallic photocathodes, ranges between 10^{-4} and 10^{-3} respectively when illuminated with 266 nm light [15, 16].

A few years ago pure Yttrium has been made a subject of photoemission studies: its work function value, which is about 3.1 eV, closely matches the photon energy available from a frequency doubled commercially available Ti:Sapphire laser. The QE of several 10^{-5} at 406 nm illumination offers an advantage over Cu allowing in principle the operation of the cathode by using the visible radiation [17].

Despite the notion that pure metals should represent a rather simple test platform for the photoemission theories it became evident by comparing the experimental results and theoretical expectations that even these systems oftentimes deliver unexpected results. Thermal emittance values measured for Cu photocathodes have been reported to be a factor of two larger than the theory predictions [18] while Mg photocathodes yielded experimental values is a factor two lower [19]. Recent characterizations on Cu photocathodes used in an RF gun showed that the surface of a real photocathode is far from the ideal one assumed in developing the photoemission models: micro-diffraction analyses revealed patches of Cu(100) and Cu(111) crystal surface orientations. Considering that the work functions for the Cu(100) and for Cu(111) surfaces are 4.59 and 4.94 eV respectively ,thus, giving different QE and MTE for the same wavelength, and the fact that the size of these domains ranges between a micron to a millimetre scale, it becomes evident how the real case can be more complex than a theoretical model used to describe it. Moreover the surface roughness obtained with profilometric measurements is not negligible either and may affect the emission process. The surface roughening has been pointed out as a possible cause of thermal emittance increase via the spread of transverse velocities of emitted electrons [20]. In addition, the presence of the rough surface causes a distortion of the extracting electric field lines that may have two effects degrading the thermal emittance: a local lowering of the work function due to the tip effect, and a non zero transverse component from the distorted electric field lines which deflect the electrons from their ideal trajectories [21].

Recent results obtained in both RF and in DC gun test chambers show that despite their claimed contamination insensitivity even in UHV (10^{-9} mbar range), both the low work function metals such as Mg and Y as well as the more inert Cu may suffer from the contamination due to chemical species present as residual gases (H_2, CO, CO_2, H_2O) leading to QE decrease on a time scale ranging from hours to months depending on the material and the system [22-24].

Laser cleaning, hydrogen ion bombardment and exposure to ozone have been successfully used to remove the surface contaminants from the surface of metallic photocathodes and to recover the QE and uniformity of emission [25-27].

While the laser cleaning procedure, performed in situ in an RF gun, has been effective in recovering the overall QE and uniformity of emission by removing the polluted layers [25,28] the associated increase in the surface roughening has not been solved [21]. Hydrogen ion bombardment has also been effective in removing the contaminants from Cu surface thought the effects of surface modification have still to be studied [26]. The exposure of Cu to ozone has been demonstrated to be effective in removing surface carbon contamination. It was found that the ozone reacts only with the surface adsorbed carbon producing CO in gas phase without oxidizing Cu [27].

The use of ultrathin protective coating of wide band gap materials that do not absorb drive laser photons is another interesting approach in find a solution against the contamination of the metallic surface. Once the surface of the metal surface is placed in contact with the thin film semiconductor, the band diagrams of the two materials align with respect to their vacuum levels. Electrons excited in the metal and injected with energies above the vacuum level travel to vacuum-solid interface through the semiconductor coating region suffering scattering mainly with phonons, which could preserve their energy at a high enough level to be extracted from the photocathode. Such configuration is very promising because in principle it allows to increase the lifetime of the metallic photocathodes by passivating the surface against adverse chemical reactions. Moreover the quantum efficiency of the metallic photocathode could be increased by antireflective properties of specially engineered film at a given operational laser wavelength.

Maldonado and collaborators [29] reported experimental results of Cu photocathode covered by a thin (~18 nm) film of CsBr. The QE increase by a factor of about 50 (at 257 nm or 4.8 eV photons) compared to the best values obtained from a clean copper surface has been demonstrated. Furthermore, the photoemission yield has not been strongly affected from a brief exposure (about a minute) to normal atmosphere air. The higher yield of such photocathodes has been attributed to the presence of intra-band states lying at energies between 3.7 and 4.0 eV lower than the conduction band minimum (CBM) of CsBr as shown by photoluminescence spectra reported by Liu and collaborators [30].

The use of a wide band gap material in form of thin film typically does not appear to contribute to significant lengthening of the response time from these photocathodes. Bunches as short as 500fs FWHM have been obtained from a Cu cathode covered with a MgF_2 thin film in the so called dynamical blowout regime in a S-band RF gun by Musumeci and collaborators [31]. While a lengthening of such short electron pulses is expected due to space charge forces acting within the bunch, the longitudinal temporal profile does not show any tails typical of long response time.

Another pure metal that is considered an attractive source of photoelectrons is Pb. The photoemission properties of Pb have been investigated in recent years primarily due to its superconducting property (below 7.2 K) and remarkably high QE for metals (~$2.7x10^{-3}$) when illuminated with UV light (213 nm) [32]. Much effort has been given to optimize the deposition of a thin Pb films by means of cathodic arc discharge directly inside the endplate of a Nb superconducting RF gun using a magnetic filter with the aim to decrease the number of droplets and debris at the film surface [33]. Preliminary measurements indicate that the presence of the Pb film only slightly affects the quality factor of the gun SRF cavity [34] but the operation of this photocathodes inside the superconducting RF gun has not yet been demonstrated.

The consequences of Cs atoms partially covering metallic surfaces have been investigated experimentally and theoretically showing a good agreement between them. Tungsten cesiated surfaces demonstrated reasonable QE values for wavelengths ranging from UV to IR [35]. Nevertheless, desorption and contamination of the surface Cs due to residual gases contribute to QE degradation. One proposed solution is in supplying freshly evaporated Cs to metal surface has been demonstrated by Montgomery and collaborators [36]. The same group is developing a theory to model the diffusion process of caesium towards the surface of a metallic photocathode through microvias generated with a suitable pattern in order to optimize the uniformity of Cs surface coverage and hence that of the photoemission [37].

Semiconducting Photocathodes

Semiconducting photocathodes offer many advantages over the metallic photocathodes with their very high values of QE as compared to metal cathodes (up to 30-40%) and often not requiring a UV laser leading to a much less stringent drive laser system. However, they require much better vacuum than metals (not exceeding 10^{-7} Pa) for production, handling, and operation.

As mentioned earlier, the semiconductor photocathodes can be divided in two main categories; the positive electron affinity (PEA) and negative electron affinity (NEA) types depending on whether their vacuum energy level lies above or below the CBM.

The most studied PEA materials are cesium telluride (Cs_2Te) and alkali antimonides. Both are commonly synthesized in forms of thin films on top of conducting substrates by using thermal evaporation in UHV (~10^{-8} Pa) growth chambers.

Cs_2Te cathodes can have QE in excess of 15% when illuminated with 254 nm photons [38].These cathodes can operate reliably on time scale of several months [39]. These thin films have proven to withstand electrical field intensity up to 60 MV/m. Applying CsBr protective coating of 2 nm thickness significantly decreased the QE, without improving the lifetime, in addition, the UV photoemission threshold was shifted from 3.9 eV to 4.1 eV [40]. Increasing the coating thickness up to 4 nm, resulted in a QE of 7% but kept the QE stable for at least 2 months [41]. During the last 15 years more than 100 photocathodes have been grown and operated in RF guns allowing collection of large amounts of experimental data that demonstrate reliable and reproducible results [42].

Production recipes of alkali antimonide photocathodes such as NaK_2Sb and CsK_2Sb have been detailed in the literature [43] and optimized during the last decades by the photomultiplier industry. QE measurements indicate that these materials are suitable to achieve quantum efficiency values exceeding 30% when illuminated with photons at about 400 nm [44].

The synthesis of these materials is quite complex, involving the use of highly reactive pure alkali metals, and require a good control of all deposition parameters during the growth, frequently resulting in photocathode characteristics that differ slightly from sample to sample. The lifetime of this type of photocathodes is very sensitive to contaminants such as oxygen and water vapour but can be extended at the expense of reduction in QE by using very thin protective coatings of CsBr, NaI or CsI [45, 46]. The limited lifetime in RF guns, due to the poor vacuum, is likely the main reason why these photocathodes have not been considered despite the fact that a CsK_2Sb photocathode holds the world record of average current produced from a photoinjector [47].

On the other hand, DC guns routinely provide outstanding vacuum levels (10^{-10} Pa) that makes possible to improve the lifetime if the degradation is only due to residual gas contaminations by orders of magnitude. The lack of experimental data for alkali antimonide photocathode in terms of performance in photoinjectors and the availability of DC photoinjectors make a strong case for an interest surge in studying these materials in connection with very high average current photoinjectors [48].

NEA surfaces in photocathodes are generally achieved using semiconductor such as GaAs, GaAsP or GaN. The procedure consists in exposing a single crystal surface to alternating fluxes of Cs and O_2 or NF_3. This will results on the formation of a surface dipole strong enough to lower the vacuum level below the conduction band minima. The procedure take the name of "yo-yo" technique based on the fact the while alternating Cs and O_2 of NF_3 the quantum efficiency will be respectively raised or lowered. The activation process is stopped once a plateau is reached in the photoelectric yield. As the NEA condition is related to only few surface monolayers, it is extremely sensitivity to contaminants requiring most stringent vacuum of 10^{-9}–10^{-10} Pa [49]. NEA cathodes also are the most sensitive to the problems of ion back bombardment caused when the electron beam impact-ionizes the residual gas in or near the DC gun [50]. Even with long and complex optimization of the beam transport, the charge extracted from a GaAs photocathode from a single laser spot does not exceed about 600 Coulomb (1/e lifetime) at a current levels of 5-10 mA [51]. Re-caesiation of the surface may be not be sufficient to fully restore the NEA and QE, although a

heat cleaning treatment restores the photocathode performance to essentially its original performance.

High temperature heating of semiconducting wafers carried out with the aim to clean the crystal surface from residual physisorbed chemical species to prepare the surface for the NEA activation may result in a roughening of the crystal surface and to a consequent unexpected increase of the thermal emittance [52, 53]. For this reason particular care should be taken in controlling the sample temperature during activation experiments.

GaAs and GaAsP have been demonstrated to be very promising sources of high brightness electron beam showing a respectable quantum efficiency of a few percent (and as high as 20%) with thermal emittance values as low as 0.12 mm-mrad/mm-rms (GaAs illuminated with 860 nm laser) and with short response time (≤0.14 ps for GaAs illuminated with 460 nm laser) [54-56]. Is is worth noting that these properties have optimal values at different wavelengths: for example at longer wavelength (860 nm) the measured response time is longer tens of ps and thermal emittance low while the shorter wavelength (460 nm) give the shortest response time. The choice of the operational laser wavelength should take into account the trade-off between these properties. A possibility of sub-thermal MTE from this photocathodes even at short wavelengths has been recently considered in [53]

The main drawback of these photocathodes still remains in their high sensitivity to the contaminants [56].

GaN has demonstrated high QE value of about 50% when illuminated by UV photons at a wavelength of 312.6 nm [57]. The thermal emittance has been evaluated to be rather high at a wavelength of 260 nm (1.35 mm−mrad/mm rms), while the emission response was very prompt without showing any tail on the current profile [58]. Quantum efficiency of the photocathode is maximized to 71.9% at photon energy of 5.4 eV by an Mg-doping concentration of 3.0×10^{19} cm^{-3} [59].

Secondary Electron Amplifier

A very interesting approach in developing a new family of hybrid photocathodes, making advantages of the properties of PEA and NEA materials, has been recently proposed and is undergoing development being based on diamond amplification trough secondary electrons [60]. Primary photoelectrons are accelerated at relatively low energy towards a thin diamond window where they could generate a large amount of secondary electrons, which relax to CBM and extracted into vacuum through a NEA surface achieved by hydrogen termination of the diamond window [61]. By combining the properties of these two classes of materials is it possible to foresee the production of very intense electron beams with very low thermal emittance.

Current amplification with gain up to 400 have been obtained within the diamond window [62] and the first electron beam has been extracted from diamond window and transported into vacuum with a gain of about 40 [63].

Others

Alloying pure metals such as Al, Mg and Cu respectively with small amounts of Li, Ba and BaO leads to a strong decrease of the workfunction of these photocathodes allowing single photon emission also with visible radiation. Within these alloy is of particular interest the optimized alloy Mg-2.1%Ba: it shows an impressive QE for metals of about 2% when illuminated with 4.9 eV photons [64].

Another possible approach to the generation of high brightness electron beam goes through the engineering of materials allowing the photoemission process to a single surface band that limits the transverse velocity spread of electrons. A proposed structure suitable to allow this process consists of alternating ultrathin layers of MgO and Ag(100). Mathematical models based on Density Functional Theory foresee that 2-3 monolayers of MgO over an ultrathin, less than 8 monolayer, Ag(100) film induces a decrease of Ag workfunction from 4.6 to 2.6 eV and electron emission is expected to have a value of 0.06 mm-mrad/mm-rms [65].

CONCLUSIONS

While during last years some progresses have been made on the understanding of fundamental processes leading to the generation of high brightness electron beam in photocathodes gun some experimental observations are not yet completely described by the theory and on the other hand there is still a lack of required experimental data to validate models predictions.

The complexity of the physical processes involved implies that further remarkable improvement of photoelectron beam properties will require expertise from multiple scientific fields to collaborate to define experimental procedures aimed at improving the performances of existing photocathodes and to characterize them providing new experimental results to be compared with theoretical expectations.

The support and collaboration of dedicated facility or laboratories is recommended with the aim of not jeopardizing experimental results and to allow suitable photocathodes candidates to be promptly tested inside RF or DC gun allowing identification of advantages and drawbacks of different materials when operated in applications with accelerator machines.

An overview of photocathodes currently used and developed with the aim of generating electron beams in photoemission guns has been presented. Attention has been focused on the state-the-art and improvements foreseen to find solutions to the physical and technological limits presented by different materials.

REFERENCES

[1] P. Castro, Proceeding of LINAC 2000, 696 (2000)
[2] R. Hajima, *Proceeding of APAC07*, 11 (2007)
[3] R. Tomás, *Phys. Rev. ST Accel. Beams*, **13**, 014801 (2010)

[4] M. Ferrario et al., *Phys. Rev. Lett.,* **104**, 054801 (2010)
[5] I.V. Bazarov, B.M. Dunham, C.K. Sinclair, *Phys. Rev. Lett.,* **102,** (2009) 104801
[6] O. J. Luiten et al., *Phys. Rev. Lett.* **93**, 094802 (2004)
[7] S. Schreiber, *Proceedings of FEL 2006*, 590 (2006)
[8] N. Yamamoto et al., *J. Appl. Phys.*, **102**, 024904 (2007)
[9] D.H. Dowell et al. , *Nucl. Instr. Meth. Phys. Res. A*, **622**, 685 (2010)
[10] D. H. Dowell and J. F. Schmerge, *Phys. Rev. ST Accel. Beams*, **12**, 074201 (2009)
[11] K. L. Jensen, *J. Appl. Phys.*, **102**, 024911 (2007)
[12] W. E. Spicer, *Phys. Rev*., **112**, 114 (1958)
[13] P. Musumeci et al., *Phys. Rev. Lett.*, **100**, 244801 (2008)
[14] M. Boscolo et al., *Proceedings of EPAC06*, 98 (2006)
[15] P. Davis et al., *Proceedings of PAC93*, 2976 (1993)
[16] T. Nakajyo, et al., *Jpn. J. Appl. Phys.*, **42**, 1470 (2003)
[17] L. Cultrera et al., *Radiat. Eff. Def. Sol.,* **165**, 609 (2010)
[18] Y. Ding et al, *Phys. Rev. Lett.,* **102**, 254801 (2009)
[19] H.J. Quian et al., *App Phys Lett*, **97**, 253504 (2010)
[20] M. Krasilinokov, Proceedings of FEL2006, 583 (2006)
[21] D.H. Dowell, *Workshop on Photocathode Physics for Photoinjectors*, Brookhaven National Laboratories (2010), http://www.bnl.gov/pppworkshop/
[22] L. Cultrera, *Workshop on Characterization of High Brightness Beams*, Zeuthen, (2008) http://www-zeuthen.desy.de/chbb08/
[23] L. Cultrera et al., *Nucl. Instr. Meth. Phys. Res. A*, **587**, 7 (2008)
[24] L. Cultrera et al., *J. Nanosci. Nanotechnol.*, **9**, 1585 (2009)
[25] L. Cultrera et al., *J. Phys. D: Appl. Phys.,* **40**, 5965 (2007)
[26] D. H. Dowell et al., *Phys. Rev. ST Accel. Beams*, **9**, 063502 (2006)
[27] G. Penco et al., *Proceedings of IPAC2010,*1293 (2010)
[28] L. Cultrera et al., *Proceedings of DIPAC2009,* 140 (2009)
[29] J. R. Maldonado et al., *Phys. Rev. ST Accel. Beams*, **11**, 060702 (2008)
[30] Z. Liu et al., *Appl. Phys. Lett*., **89**, 111114 (2006)
[31] P. Musumeci et al., *Phys. Rev. Lett.,* **104** 084801 (2010)
[32] J. Smedley et al., *Phys. Rev. ST Accel. Beams*, **11**, 013502 (2008)
[33] P. Strzyzewski et al., *Proceeding of EPAC06,* 3209 (2006)
[34] J. Sekutowicz et al., *Proceeding of EPAC06,* 3493 (2006)
[35] N. A. Moody et al., *Appl. Phys. Lett*., **90**, 114108 (2007)
[36] E. J. Montgomery et al., Proceeding of Advanced Accelerator Concepts: 13th Workshop, 599 (2009)
[37] E. J. Montgomery et al., *Journal of Directed Energy*, **3**, 66 (2008)
[38] L. Monaco et al., *Proceedings of PAC07*, 2763 (2007)
[39] P. Michelato et al., *Proceeding of EPAC08*, 241 (2008)
[40] E. Chevallay et al., *Proceeding of LINAC 2000*, 110 (2000)
[41] D. C. Nguyen et al., Nucl. Instr. and Meth. in Phys. Res. A, **429**, 125 (1999)
[42] http://wwwlasa.mi.infn.it/ttfcathodes/
[43] E. Shefer, A. Breskin, A. Buzulutskov, R. Chechik, M. Klin and M. Prager, *Nucl. Instr. and Meth. in Phys. Res. A,* **411**, 383 (1998)
[44] R. Mirzoyan et al., Nucl. Instr. and Meth. in Phys. Res. A, **567**, 230 (2006)
[45] A. Buzulutskov et al., Nucl. Instr. and Meth. in Phys. Res. A, **400**, 173 (1997)
[46] E. Shefer et al., Nucl. Instr. and Meth. in Phys. Res. A, **433**, 502 (1999)
[47] D.H. Dowell et al. , *Nucl. Instr. Meth. Phys. Res. A*, **356**, 167 (1995)
[48] L. Cultrera et al., WEP244, these proceedings
[49] J. Qiao et al., *Proc. SPIE 6621*, 66210K (2007)
[50] C. K. Sinclair et al., *Phys. Rev. ST Accel. Beams* **10**, 023501 (2007)
[51] T. Siggins et al., *Nucl. Instr. and Meth. A* **475** 549 (2001)
[52] S. Karkare et al., TPH244, these proceedings
[53] S. Karkare and I. Bazarov, *Appl. Phys. Lett*., **98** (2011) 094104
[54] I. V. Bazarov et al., *J. Appl. Phys.,* **103**, 054901 (2008)
[55] I. V. Bazarov et al., *Phys. Rev. ST Accel. Beams*, **11**, 040702 (2008)
[56] J. Grames et al., *Proceeding of PAC05,* 2875 (2005)
[57] F. Machuca et al, *J. Vac. Sci. Technol. B.*, **18**, 3042 (2000)
[58] I. V. Bazarov et al., *J. Appl. Phys.,* **105**, 083715 (2009)
[59] S. Uchiyama et al., *Appl. Phys. Lett*., **86**, 103511 (2005)
[60] I. Ben-Zvi et al.,"Secondary Emission Enhanced Photoinjector", BNL Report CA/AP/# 149
[61] T. Rao et al., Proceeding of Advanced Accelerator Concepts: 11th Workshop, 178 (2004)
[62] X. Chang et al., *Proceedings of PAC07*, 2044 (2007)
[63] X. Chang et al., *Phys. Rev. Lett.,* **105**, 164801 (2010)
[64] V.G. Tkachenko et al., *Appl Phys B*, **98**, 839 (2010)
[65] K. Németh et.al, *Phys. Rev. Lett.*,**104**, 046801 (2010)