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REVIEW OF RECENT PHOTOCATHODE ADVANCEMENTS

L.Monaco* and D. Sertore, INFN Milano - LASA, Segrate, Italy

Abstract

Photocathodes are routinely used as a source of electrons in high brightness beam photoinjectors. The properties of the photocathode have a significant influence on the parameters of the electron beams and on the operation of the machines. The choice of photocathode materials is an important step in reaching the challenging requirements of modern accelerators. Recent advancements towards more performing photocathodes are here presented and discussed.

INTRODUCTION

Photocathodes are key components of modern and advanced high brightness electron sources [1]. Illuminated by a laser beam, photocathodes emit electrons at proper phases in a high electric field region, necessary to accelerate the electron bunch to high energy to compensate for space charge forces and optimize beam emittances.

To generate the required electric field, the most common solutions are DC and RF guns. The solution adopted by different labs is mainly related to the kind of application of the specific machine. Indeed, polarized electron sources typically use DC guns due the extreme requirements of vacuum necessary for the operation of these kind of photocathodes (i.e. GaAs/Cs). Nearly the same vacuum conditions are necessary for the operation of alkali antimonide photocathodes due to their sensitiveness to vacuum level and composition. Recently, antimonide photocathodes have been used also in RF guns with limited performances due to dark current and limited operative lifetime. RF guns are instead routinely used with metallic photocathodes and, in case of high QE (Quantum Efficiency) materials, with cesium telluride (Cs_2Te).

Photocathodes properties determine also the minimal electron beam emittance through the so called thermal emittance. This property depends on the electron transverse velocity and hence on many parameters like photocathode surface roughness, distribution of work function on the surface, etc. This has motivated, in the present years, a strong activity based on solid state physics, surface science and material engineering to improve and optimize present photocathodes and explore new materials.

This paper presents the recent photocathode advancements with emphases on semiconductor photocathodes and their application in user dedicated accelerators. These materials are nowadays used in the main FEL user facilities and they are the best candidates for the coming new advanced CW FEL machines (see LCLS-II, SHINE, ..) and very high current electron source for ERLs or for the beam cooling of the Electron Beam Collider (EIC).

* laura.monaco@mi.infn.it

PHOTOCATHODE REQUIREMENTS

Photocathodes requirements are specific to their applications and a simple classification is not easily achievable.

However, it is possible to identify a common set of properties that are a minimal requirement for application of photocathode in modern accelerator machines [2]. They can be summarized in the following points:

- Long operative lifetime. This is important for photocathode operation in user facility based machine. Long operative lifetime implies also stable photocathodes properties in particular for operation in RF guns.
- Low dark current. Activation of components along the accelerators must be avoided and, hence, it is important to limit field emitters both from the photocathode and from the gun. In DC gun operation where the electric field is static, this is even detrimental for the photocathode itself since it generates ion back bombardment that damages the photoemissive film.
- Fast response time. Prompt response of the photocathode material to laser illumination is mandatory to guarantee proper phasing between laser and electric field (in particular for the RF gun operation). Moreover, the emitted electron bunch needs to be as similar as possible to the laser beam profile in order to minimize the effect induced by the space charge on the electron beam emittance.
- High QE. This parameter is extremely important for high repetition rate or CW applications. Lower repetition rate electron sources usually use metal photocathodes with QE in the 1×10^{-6} range.

Besides the previous presented parameters, electron polarization is a very specific requirement that only a family of photocathode is able to satisfy, i.e. III-V materials. This class of photocathodes is the subject of many studies towards improving the polarization ratio and the lifetime. Indeed these are Negative Electron Affinity (NEA) photocathodes and they need usually an atomic layer of Cesium on the surface to preserve the electron polarization. Consequently, the specification for vacuum level and composition are very demanding and require specific vacuum system. It is then clear that polarized photocathode are a class of photocathodes by itself and, given the limited space here available, they will not be addressed it here but recent updates have been presented at the Snowmass2021 Electron Workshop [3].

METAL

Metal photocathodes are widely used in low repetition rate electron sources and where fast response time and very low emittance are required.

The most common material is copper (see LCLS for example) but, recently, magnesium has been used in SRF gun at HZDR Elbe [4–6]. Given the “simplicity” of the mate-

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rial and the possibility to machine and then use it without particular regards about keeping vacuum conditions, there has been many proposal for surface topological engineering to enhance the photocathode performance. Recently, it has been proposed a photocathode with spiral flat microemitters machined on the exposed surface to enhance the emitted electron beam brightness [7].

Being the QE for metal low, several attempts are pursued to increase it. This is typically done by depositing very thin layer of properly selected materials on top of the photocathodes in order to lower the work function of the composite structure. This has been done at STFC by depositing a thin layer of MgO on top of a copper cathode [8], showing a significant improvement of the photocathode QE. Concerning the robustness, at STFC a comparative study of the QE degradation of polycrystalline and single-crystal Ag samples induced by oxygen shows that the effect is more relevant on single-crystal and, that can be partially recovered with the exposition of carbon monoxide. MTE (Mean Transverse Energy) measurements at room and cryogenic temperature reveal that QE decrease is more rapid at low temperature due to the increased sticking probability of the oxygen on the photocathode surface [9].

ALKALI TELLURIDE AND ANTIMONIDE

Alkali telluride and antimonide photocathodes nowadays are the most used materials for delivering high brightness electron beam.

The choice of these materials is mainly based on the need of having high QE photocathodes for high repetition rate and/or high average current applications. While alkali telluride provide reliable and stable performances, they need UV light (4th harmonic of common lasers), a drawback that the alkali antimonide try to solve being sensitive to visible light and hence needing only the second harmonic, so relaxing request on the light sources. Both require a high quality vacuum, more stringent for the antimonide ones that result to be less robust than the telluride ones. The vacuum level required are of the order of 1×10^{-11} hPa and 1×10^{-10} hPa, for alkali antimonide and telluride respectively. Here below, the two families (anitmonide and telluride) of photocathodes will be shortly presented with focus on recent advancements.

Cs_2Te

Cs_2Te is commonly used in high repetition rate accelerator due to its high QE, long operative lifetime, relatively low emittance and fast response time. These performances are the result of more than twenty years of development starting from the production, operation in RF guns and post-operation analysis done at INFN Milano - LASA [10].

These photocathodes are used as electron source in user facilities like FLASH (DESY), European XFEL (Germany), LCLS-II (SLAC, USA) and in many accelerators around the world. The performances of cesium telluride are now taken as reference for the development of new photocathodes. Just to give some number: the QE at the production is in 10 % range and it drops during operation to few percent; operative lifetime is now around 1400 d; response time in the hundredths femtosecond range [11]; thermal emittance below 1 mm/mrad; dark current well below limiting values [12–16].

Recently HDZR-ELBE join the list of laboratories that use cesium telluride photocathodes. They successfully deposited Cs_2Te on copper substrate [17] to improve thermal conductivity with respect to Mo. Since it start operation in May 2020, seven films have been produced and operated in CW condition up to 100 kHz, with an average lifetime between 2 and 3 months. The total extracted charge, from Jun 2020 to Feb 2022, has been 91.3 C for a total of 3064 h of user beam.

Co-evaporation towards production The improvement of thermal emittance is based on the pioneering work of Gaowei [18] that showed how the adoption of a co-deposition process for the film growth reduces significantly the roughness and hence its contribution to the photocathode thermal emittance. Since then, many labs are working towards the implementation of this technique into photocathode growth.

A collaboration between CERN and ASTeC has recently grew four photocathodes by coevaporation on copper substrates at CERN. The samples were transferred to STFC where they were fully characterized by QE, XPS and TESS. These studies showed a large variability in the performance of the six photocathodes. The authors measured also Mean Transverse Energy (MTE) at different temperatures from 178 K to room temperature. The MTE curves change with temperature as expected. Authors measured, on one of the photocathodes, an increased of transverse energy above 400 nm (below photoemission threshold), probably due to surface compounds with a low surface coverage and therefore low effective QE [19].

ASTeC has also started an its own activity dedicated to cesium telluride in the framework of CLARA activities. Interestingly, they have grown two photocathodes on Mo: one with sequential deposition and one with coevaporation. They use ion beam sputtering for Te evaporation. Their XPS analysis shows in both cases mixed phase of Cs_xTe and, in the case of coevaporation, some amount of oxygen probably coming from the Cs source, not yet fully conditioned [20].

Alkali antimonide (Na-K-Cs-Sb)

Cesium telluride, although its good performance, has the drawback of being photo sensitive only to UV light. This has some implications one which is the need of the forth harmonic of the fundamental wavelength of common lasers to operate. Despite the high QE, this make the cesium telluride impractical for very high repetition rate or high average current operations. The natural choice is to move to alkali antimonide that are sensitive to visible light and hence require only a second harmonic to photo emit. Their sensitivity to vacuum conditions however requires at least an order of magnitude better vacuum with respect to telluride photocathode.

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The multi alkali antimonide have gain a new interest in the recent years supported by an intense R&D activity for the improvement of their properties in term of robustness (and lifetime) and thermal emittance. Moreover, they have benefit from the improvement on the vacuum technology that allows their operation not only in DC but also in RF guns.

DC gun operation Electron-Ion Colliders (EIC) ask for unpolarized electron beams with high average current, low emittance and long operative lifetime (I > 100 mA, $\epsilon \approx 1.5\,\mathrm{mm\,mrad^{-1}}$, > 3 days) [1] for producing cold electron beams for hadron cooling. Thanks to their capability to work in visible range providing high QE and their low emittance, antimonide photocathodes are nowadays the best candidates. The operation of K-Cs-Sb in DC guns has been demonstrated [21, 22] and up to 65 mA have been extracted [23]. These experiments show that main limitations come from the QE degradation and the short operative lifetime and these motivate several research activities dedicated to improve the photocathode performances also in view of the new advance CW FEL machines and very high current electron source for ERLs.

Main causes of QE limitation are the thermal heating and the ion back bombardment issue. It has been seen that cathode temperature and laser power shining on the coating can cause the QE drop. Indeed, QE drops above 70 °C as well as if the power of the impinging laser exceeds some W [24] . To address the above thermal limitations, different solutions are being explored that imply cooling of the cathodes (p. es. used in Cornell and SBU-BNL DC guns) [24] or the solution developed for the MIST DC gun (10 mA unpolarized beam current) for the MESA accelerator in Mainz where the heat will be transferred from the puck that hold the cathode towards the boron nitride (BN) stem via a metallic gripper [25].

The ion back bombardment on the cathode surface caused by the beam ionized vacuum gases has been avoided by depositing off center cathodes and adjusting the beam optics to compensate for this.

RF gun operation at high gradient Operating alkali antimonide photocathodes in RF gun benefits from the higher accelerating gradient available but poses new challenges on vacuum level in the RF gun and the photocathode operation in high fields.

In the framework of a INFN Milano LASA - DESY PITZ collaboration, three KCsSb photocathodes have been prepared in a new dedicated deposition system at LASA and shipped to PITZ for testing in the RF gun [26]. The sequential deposition process was developed in the R&D system at LASA and transfer to the deposition system [27]. The three photocathodes, with QE above 5 % at 516 nm, have been successfully transfer to PITZ preserving the high QE. Their operation in the RF gun has been limited to 30–40 MV/m due to vacuum events that degraded significantly the QE. The dark current of these cathodes was higher than the one measured with Cs_2Te photocathodes, mainly due to the lower photoemission threshold. At PITZ, also the response time of a low (0.4 %) QE cathode was measured to be less than 100 fs. Since the QE degradation could indicate possible changes in the surface chemistry of the film, a new measurement on a fresh photocathode must be done to confirm this preliminary results. Finally, thermal emittance and emittance measurements confirm value smaller than the ones obtained for Cs_2Te (at 40 MV/m and 100 pC about 23 % decrease of emittance with a 4D brilliance enhancement of about 60 %), consistent with expectations. Post-usage analysis highlighted the increase of the photoemission threshold and showed the appearance of a shoulder at low energy in the spectral response for one of the cathodes, probably due to oxidation of the film.

A joint project between Cornell and UCLA allowed to test a Na-K-Sb photocathode, produced at Cornell, in the high gradient gun of the PEGASUS facility at UCLA. The QE, after deposition at the Bright Beams Center in Cornell, was about 1.5 % at 532 nm. The photocathode was transported then to UCLA in a dedicated suitecase but, at its arrival, the QE was dropped to about 8×10^{-2} % at 405 nm. Despite this low value, the QE remained stable for multiple weeks of operations. The photocathode QE was measured in the RF gun as 0.5 % at 266 nm, 8×10^{-2} % at 405 nm and they also performed measurement at 800 nm showing a mixing of the 2-photon and 3-photon photoemission. Finally, the thermal emittance measurements scale consistently with the change of photon wavelength [28].

Photocathode development To improve the performance of alkali photocathodes, several researches start aiming to develop more robust alkali antimonide photocathodes (for example Na-K-Sb that can withstand higher temperatures), new protective coatings, different growing procedures to increase the final smoothness of the coating, QE increase to limit the laser power needed.

The research on multi-alkali photocathodes involves many laboratory. Recently, at HZB they have deposited NaKSb co-evaporated (Sb + alkalis coevaporation) and sequential NaKSb photocathodes, aiming of setting up the growing process towards application for their SRF gun. The QE was between 0.3–2.0 % at 515 nm. Also the lifetimes were quite widespread. XPS analysis showed indeed different compositions of the grown films. Further studies are planned to improve the reproducibility of the growing process [29].

The transfer of the growing process to substrate like the plugs compatible with RF guns is a key element for photocahode operations. This activity has been pursued in China where different laboratories have developed their own photocathode deposition system, suitcase and transfer system inspired to the INFN LASA ones [30]. Concerning photocathode deposition, at SARI, for example, they deposit firstly Sb and then they coevaporate K and Cs. During the deposition they monitor not only photocurrent but also the reflectivity to have better control of the growing process.

Brightness improvement As important as the development of robust photocathodes is the R&D activity dedicated

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to improve the brightness of the electron beam generated from the photocathodes. Sources of increase of the thermal emittance are the roughness and the non uniformity of the surface photoemission threshold. Both effects contribute to the electron transverse momentum and hence to the emittance.

To reduce these contribution, we have already mention the development of co-evaporation deposition that guarantees a smoother final surface. A further step in this direction is the result obtained by Parzick et al. [31] that succeeded in growing an epitaxial Cs_3Sb photocathodes on a 3C SiC(100) substrate. Besides already important result of a larger lattice order of the epitaxial growth photocathode, the authors achieved a remarkable QE of 2 % at 532 nm on a film thinner than 10 nm. Moreover, they observed an enhanced QE at 650 nm with respect to the standard spectral response of cesium antimonide. This epitaxial film have allowed also ARPES measurements to unveil the band structure that has been compared with DFT (Density Functional Theory) calculation. The difference between the theoretical expectation and the measured data are attributed to strain and intrinsic instability of the grown Cs_3Sb.

To further improve the smoothness of cesium antimonide photocathodes, recently a substrate of a single crystal strontium titanate (STO), that matches the lattice constant of Cs_3Sb [32], has been used to growth this photocathode. The growth on a matched-lattice has improved the smoothness and also the uniformity of the surface photoemission threshold, opening a new path towards film optimization for improved high brightness beams. STO(100) was chosen from an open-source list of 150 commercially available candidate substrates, 23 which are lattice-matched to Cs_3Sb using the MPInterfaces [33] software package (a machine learning approach).

Graphene protective coating To reduce alkali antimonide sensitivity to vacuum conditions, an interesting approach is based on layers of graphene.

A collaboration between LANL and KEK has developed a process to deposit Cs_2KSb on graphene layers [34, 35]. The authors has shown that the quality of the graphene plays an important role in the final film performances. The CsK_2Sb photocathode deposited on the proper graphene layer show good QE but also the measurement through the graphene layers present a reasonable QE, dependent on the number of graphene layers. During this study, it was observed an unexpected QE enhancement when the graphene was deposited on a metal substrate as well as that the graphene layer can be cleaned by heating above 500 °C and used as a reusable substrate for these bialkali antimonide photocathodes.

A different approach to the use of graphene layers has been presented in [36]. In this work, a 10 nm Sb layer is deposited on a Si substrate. The Sb layer is then cover with graphene multilayers. The advantage of this solution is that is possible to retain the Sb layer on the Si substrate up to 600 °C with a clear improvement on the substrate cleanliness and Sb reusability. The authors then succeeded to deposit cesium on top of this support and grow a Cs_3Sb photocathode as reported from XPS measurements. The future plan of this research is to fully characterize the photoemissive properties of this film.

QE enhancer As a last topic in this overview of new possible ideas to enhance multialkali antimonide, the proposal of using Surface Plasmon Polaritons (SPP) needs to be presented. As already done in metals, proper engineering of the photocathode substrate might be used to enhance the QE. In the approach followed by [37], the complex constituted by substrate and bialkali antimonide are designed to enhance absorption by exciting SPP at the desired photon wavelength. This model is then implemented in a Monte Carlo model and, from their simulations, the authors show a factor at least of 2 on the QE when illuminated with the selected photon energy.

PHOTOCATHODE DIAGNOSTIC IN PRODUCTION SYSTEMS

Diagnostic during the deposition of photocathode film that will be operated in electron sources in accelerator facility is important. Indeed, it allows having many important information: feedback on the deposited cathode with the possibility to improve their reliability; post usage analysis to correlate photocathode performance in operation with the measured properties; continuous improvement and R&D on cathode for accelerator applications that might behave differently from the research films. Many production systems are already equipped with interesting diagnostic and here some of the new improvements are presented.

Recently, INFN LASA [38] is developing a TRAnsverse Momentum Measurement (TRAMM) device that, when completed and fully characterized, will be installed on one of the production system present in the laboratory. This device is supported by INFN-CSN5.

At DESY, a new laboratory dedicated to photocathode is being developed [39]. Blue lab will be a deposition chamber equipped with AES, XPS and, in the future, with a electron momentum spectrometer.

Finally, the Alkali-metal Photocathode Preparation Facility (APPF) [40] has been developed at STFC. This facility has a multiprobe system (for QE and work function measurement), the Transverse Energy Spread Spectrometer (TESS) for MTE measurement and a CMA for AES investigations. A feature of this system is the ability to accept both Omicron sample as INFN type plugs allowing to cross check R&D and production photocathode in the same system.

PHOTOCATHODE TEST FACILITIES

An essential tool for developing new photocathodes and improving the present ones are dedicated test facilities able to make a full characterization of the photoemissive properties of the films as under operation. In fact, the availability of user machines is limited for obvious reasons; moreover test of new photocathodes may require a change of machine parameters to fit the experiment or can imply some risks of pollution or damages that must be avoided.

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Being so important, the most recent photocathode test facilities are here reported.

HERACLES [41] at Cornell is a facility dedicated to study photocathode lifetime while in mA operations. The facility is based on a DC gun capable to reach 200 kV and 10 mA. Photocathodes are transported by a UHV suitcase. The first photocathode tested was a cesium antimonide that delivered 8 mA beam with stable current over a 3 h period.

At INFN LASA, in the framework of the BriXSinO activities, a new laboratory is being setting up for photocathode stress test [42]. This facility is based on a DC gun designed for 100 kV and 5 mA. The photocathode under test will be Cs_2Te operated at 92.857 MHz.

Finally, recently the PITZ facility at DESY has been upgraded to allow test of of visible sensitive photocathodes. This has allowed to measure INFN LASA photocathode in 2021 and new tests are foreseen either at the end of 2022 or beginning 2023.

MODERN APPROACH TO PHOTOCATHODE DEVELOPMENT

The "art" of photocathode deposition in the recent years is moving from an empirical "trial and error" approach to a more coherent and coordinated activity that involves different disciplines. This is well represented by ACERT that aims to develop new techniques and applied to photocathodes motivated by theory and guided by real-time *in-situ x-ray* analysis [43]. The same approach is being followed also by HZB in the context of the Sealab/BerlinPRO where they developed an integrated complex able to combine theoretical investigations, surface science techniques, and deposition dedicated system [29].

Moreover, Machine Learning (ML) is coming to the photocathode selection process through the work reported in [44]. This approach is based on coupling Density Functional Theory with ML. A general photoemission model was developed and used with the ML to predict work functions of photocathodes. Ones the materials were selected, they were further screened to select based on desired criteria such as commercial availability or, for example, air stable visible sensitive photocathodes. The next step will be to experimentally verified these findings.

Not only ML has been used for photocathode selection but also Artificial Intelligence (AI) has been introduced for photocathode growth. In fact in [45] has been developed and implemented an automatic process for growth of Cs_3Sb photocathodes. This has been possible thanks to a proper selection of significant parameters to follow during the growth. This process will be extended to other photocathodes (like Cs_2Te) and the feedback loop used, now implemented, will be replaced by AI process.

CONCLUSION

This paper presents a selection, based on our knowledge, of new advancements on photocathodes. The field of photocathode is expanding very fast and we hope to have given a taste of its complexity but also of its richness and multidisciplinarity that makes it very stimulating and challenging.

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