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ADVANCES IN PHOTOCATHODES FOR ACCELERATORS*

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Abstract

This talk reviews advances in photocathode technology for accelerators: cathodes demonstrating record average currents and deliverable charge, possessing ultra-low intrinsic emittance and sub-picosecond response time. It addresses the grand challenge to combine all these useful properties into a single photoemitter - one that is being actively pursued by the research community.

INTRODUCTION

The quest for photocathodes allowing production of electron beams with increased brightness to drive X-Free Electron Lasers (X-FEL) [1], Energy Recovery Linacs (ERL) [2], electron cooling of hadron beams [3], inverse Compton scattering [4] and Ultrafast Electron Diffraction (UED) [5] experiments has in the recent years received much attention from the scientific community resulting in a stronger interaction between accelerator and solid state physicists looking to identify suitable materials with improved performances for future accelerator machines and novel applications [6].

From the point of view of the electron source for an accelerator device, the four relevant photocathode properties are: Quantum Efficiency (QE), thermal emittance or Mean Transverse Energy (MTE), response time, and the photocathode longevity or the lifetime. QE is important when defining the specifications for the drive laser: the greater is the QE the smaller is the laser power needed to extract the same current. For some application like ERLs [2] and electron cooling [3] requiring up to several hundreds of mA of average current, the QEs of few percent or more in the visible range of the spectrum are needed in order to maintain the average laser power within a few tens of Watts [2]. For some other application like single pass FELs the required average currents are usually in the range of few hundreds of micro ampere [1] and for UED the number of electron in the bunch should be kept to a minimum to avoid space charge emittance degradation and the requirements for high QE are therefore not as critical [5].

Thermal emittance and Mean Transverse Energy are the figures of merit used to estimate the angular spread of electron velocities at the cathode surface. The intrinsic emittance of the beam from a photocathode is determined by the laser beam size and the mean transverse energy (MTE) of the emitted electrons through the relation:

$$\varepsilon_{n,x} = \sigma_{l,x}\sqrt{\frac{MTE}{m_e c^2}} \qquad (1)$$

where $\varepsilon_{n,x}$ is the normalized transverse emittance in the x plane, $\sigma_{l,x}$ is the rms laser spot size, m_e is the electron mass, and c is the speed of light.

Improved performances in photoinjectors technology and in emittance growth compensation techniques result in electron beam emittances, which in the state-of-the-art photoinjectors are nowadays largely dominated by the thermal emittance at the cathode [7,8]. Thus, any improvements in lowering the MTE at the cathode surface will have a measurable effect on the transverse emittance of the relativistic electron beam. The photocathode response time is generally a trade-off determined by the absorption length of the photons, the drift velocity, and the mean free path of excited electrons. For bulk semiconductors, like GaAs activated to NEA, the response time is strongly dependent on the photon energy resulting in very short (much less than a picosecond) response in the UV and a very long (tens of picoseconds) tail in the infrared part of the spectrum [9]. For applications in the RF based accelerators, it is mandatory to operate photocathodes with response time not longer than few picoseconds to avoid long tails in the electron beam bunches. Lifetime is a measure of the ruggedness shown by the cathode to the operating conditions. Some photocathode materials like metals can survive vacuum levels of 10^{-9} Torr for months [6] while others like NEA activated GaAs can survive only a few days at vacuum levels of 10^{-12} Torr [6]. Deliverable amount of charge is another measure of photocathode longevity, which is important for high average current machines like ERLs. For an ERL based X-ray facility operating at an average current of 100mA, the cathode should deliver several thousand Coulombs per day. The same amount of charge delivered will require more than a year of operation for a lower repetition rate photoinjector generating average currents in the range of hundreds of micro Ampere. As of today no photocathode material is able to fulfill all the requirements in terms of high quantum efficiency, low MTE, short response time and very long lifetime and a trade-off between the various photocathode performances is necessary in order to select an acceptable material for a specific application.

PHOTOCATHODES

The last decade has seen an increased collaboration between accelerator and solid state physics scientific community. Several dedicated workshops have been the venue to discuss questions related to this field and to propose new approaches to improve properties of photoemitting materials [7]

Plasmonic Photocathodes

In 2008 it was proposed that an ellipsoidal electron bunch with a uniform charge density can be used to

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minimize the contribution of non-linear space charge induced emittance growth [8]. The generation of such an electron bunch can be achieved in the so called "blowout" regime by illuminating the photocathode with a very short light pulse (tens of fs) as demonstrated for the first time few years later [9]. When using very short laser pulses there exists a threshold in terms of laser power density delivered to the cathode surface above which the multi photon photoemission has been also shown to be more efficient then the process involving a single photon using a Cu photocathode illuminated by 800 nm light [10]. At this wavelength the Cu cathode reflectivity is quite large and measured to be 85%. In order to improve the coupling of photons with the metal surface a nanostructured patterning of the surface can be used [11, 12]. An grid of nanosized holes drilled using focused ion beam into the surface of a Cu sample have demonstrated to decrease the reflectivity of 800 nm photons to about 20%, this reduction coupled with an large electric field increase near the edge of the holes was used to improve the efficiency of the 3 photon photoemission process by two orders of magnitude. On the other hand, the resulting thermal emittance of the beam resulted increased by a 50% [13].

Copper with Insulating Coating

Another possibility in improving the efficiency of a Cu photocathode has been demonstrated that can be achieved through a very thin (few nm) coating of the metal surface by large band gap insulating materials. The emission improvement is achieved by a large decrease of the metal work function induced by decrease of natural electron spill-over and dipole formation at the interface of metal-insulator as described in ref [14]. In addition to that photoemission from intraband states in the insulating coating can contribute to the final measured yield [15].

KBr, CsBr and CsI have been used to coat Cu samples and have shown to improve the emission yield at 266 nm with respect to the uncoated sample respectively by a factor 2.6, 77 and 2700 [16, 17]. At this time no experiment have been performed aimed at measuring thermal emittance and response time properties of these coated metallic cathodes.

Dispenser Cathode

The dispenser cathode idea resides on the measured workfunction lowering induced on tungsten when its surface is covered with a sub-monolayer of Cs atoms. In order to refurbish the W surface with Cs atoms to compensate for evaporation losses a suitable dispenser is operated on the back of the cathode to release Cs vapors that, migrating through pores into the W structure, reach the surface to keep its fractional coverage to the optimal point (measured to be 0.55 monolayer) to maximize the emission efficiency ($3x10^{-4}$ at 532 nm). In order to improve the uniformity of emission the emitting structure consists of a bunch of thin (about 20 micron diameter) W wire pressed and sintered together. This structure present a well ordered array of microchannel used by the Cs atoms to migrate to the surface. When operating at moderate temperature (125 C) this photocathode present a very uniform QE over its surface (less than 1% variation) and QE result stable within a few percent for an estimated lifetime of the dispenser of more than 30000 hours [18].

Multilayers

An example on how the solid state physics community is collaborating with the accelerator community comes from the Ag(001)/MgO photocathode. By using numerical simulations typical of solid state physics it has been estimated that the band diagram of a structure consisting of 2 or 3 monolayer of MgO epitaxially grown on both sides of 4 monolayers of Ag(001) it such that the emitted electron could have a minimized transverse momentum spread resulting in beam emittance that can be as low as 0.06 mm mrad/ mm rms [19]. A first approach to realize such a structure and to measure the emission properties has been recently reported [20]. The structure realized is not multilayer but consists of few MgO layers epitaxially grown over Ag(001) The number of layers is not uniform over the surface because of the natural tendency of MgO to grow as 2D islands. The thickness of the MgO layer was estimated to be 4 monolayers on average. This number of monolayer does not optimize the reduction of transverse momentum spread and by integrating the UV angle resolved photoelectron emission spectrum a thermal emittance of 0.97 mm mrad/ mm rms it has been measured. In addition the emission efficiency of the structure improved by a factor 7 the emission of the Ag(001) surface. All measurements have been carried out using light at 266 nm.

Caesium Auride

Already in 1943, Sommer [21] reported on the reaction of gold with alkali metals which yielded new semiconductors materials showing photoemission in the visible range. On the other hand, the measured quantum efficiencies of these new materials were so low that a practical use was ruled out. Recently, new experiments focused on maximizing the electron yield instead of the compound stoichiometry, which were aimed by initial Sommer's experiments, showed that QEs up to percent level can be achieved in the visible range of the spectrum from CsAu [22]. This material also proved to be rather resistant under exposure to moderate temperatures showing a 1/e lifetime of few ten of hours.

Alkali Antimonide

Alkali antimonide photocathodes have been demonstrating very high QEs in the visible range. Typically 10% QEs can be achieved at 532 nm [23]. Thermal emittances have been measured during the last few years for Cs_3Sb, CsK_2Sb and Na_2KSb at different wavelength in the visible range. At 532 nm thermal emittances of 0.56 mm mrad/mm rms have been measured for Cs_3Sb [24] and CsK_2Sb [25] and 0.47 mm mrad/mm rms for Na_2KSb [26] using the solenoid scan technique in a high voltage DC gun. Response time when

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excited with green light has been measured using a deflecting cavity to be not longer than a ps [24,25,26]. As of today the alkali antimonide photocathodes held the record in terms of average photocurrent ever generated by a photoinjector. Cs_3Sb was used to deliver as high as 33 mA of average current and showed no QE decay during 4 hours of continuous operation delivering more than 500 Coulomb [27]. CsK_2Sb photocathode delivered up to 65 mA of average current. The longest time span of continuous operation was of about 25 minute but this was due to frequent interruptions of operation due to vacuum events in the RF couplers of accelerating cavities used to boost the energy of the beam to few MeV. From the measured QE decay 1/e lifetime of about 30 hours was estimated and total charge in excess of 2000 Coulomb was exctracted from a single spot [27]. Na_2KSb showed similar performances delivering more than 2000 Coulomb from a single spot at a current level of 65 mA with a 1/e lifetime estimated to be 66 hours [28].

It has been pointed out that depending on the recipe used to synthesize bialklai antimonide photocathode the surface roughness induced emittance growth can be so large to avoid operation of these photocathodes in gradients of few tens of MVolt/m [29]. The surface of cathodes grown with the recipe used in photomultipliers photocathodes observed with an atomic force microscope is characterized by columnar structures with roughness on the range of 25 nm rms and periodicity of about 100 nm [30]. In order to understand better why the columnar structure formation dedicated experiment of bialklai antimonide growth have been performed while looking in situ at the structural properties of the films using X-ray diffraction and reflection. Results indicate that during the first stage the crystal structure of Sb film is dissolved during evaporation of K and replaced by the K_3Sb structure. Finally, the addition of Cs while replacing the K does not change the type of crystal structure but rather act to increase the volume of the unit cell [31]. This indicates that recipes used for growing this class of materials needs to be improved to yield smoother surfaces compatible with operation in high fields.

An extension of the tree step model used to estimate QEs and thermal emittance on metallic photocathodes that includes also the finite temperature of Fermi-Dirac distribution indicates that a limit to the minimum thermal emittance of electron beam exists and coincides with an MTE of electrons equal to kT, where k is Boltzmann constant and T the temperature of the cathode [32]. As long as the photo excited electron are coming from the tail of the Fermi-Dirac distribution the same formula applies also to semiconductor materials. Already in 1958 Spicer reported on the existence of filled donor states within the band gap of alkali antimonide materials [33]. A CsK_2Sb excited with 690 nm laser wavelength was used to generate electron beam at 300 K and 90 K using a small high voltage DC gun with a maximum field of 2 MVolt/m. As expected the QEs at cryogenic temperature was lower than that at room temperature ($5x10^{-5}$ and $2x10^{-4}$ respectively) because the density of populated states strongly depended on the temperature for this range of energy. Accordingly to the recent proposed model electron beam MTEs of 22 meV and 13 meV have been measured at 300 K and 90 K, those values agree quite well with the expected values of 25 meV and 9 meV [34].

For comparison the Cu photocathode operating at LCLS has a typical QE of $1x10^{-4}$ and MTE of 413 meV when operated at the 253 nm laser wavelength [35]. Subthermal electron beam generation has been demonstrated using photoionization of cold Rb atom, but due to practical limitation the bunch charge is limited to a fraction of femto Coulomb [36].

GaAs

The discrepancies between the expected and measured MTEs as function of wavelength called to the need of a better understanding of the relevance of the different physical process involved during the excitation, transport and emission of electron from negative affinity surface of GaAs photocathodes. Detailed numerical simulation using Monte Carlo techniques demonstrate a substantial agreement between simulation and experiments with the only free parameter in simulation being the electron affinity at the surface [37]. The code initially developed for bulk material was successively improved with the capability of handling multi-layered structure with different doping density [38]. Numerical simulations indicated that a 100 nm layer of intrinsic GaAs grown over the surface of a p-doped ($5x10^{18}$ cm^{-3}) can improve the relaxation process electrons during the transport towards the surface to vacuum interface leading to the production of electron beam with reduced thermal emittance. Experiments were carried out to compare MBE grown samples with and without the intrinsic layer. A substantial agreement with the numerical simulation was observed and a reduction of photoelectron beam MTEs by a 50% (from 120 to 80 meV, corresponding to 0.4 and 0.5 mm mrad/ mm rms thermal emittances) was measured when illuminating the photocathode surface with 532 nm wavelength. Improving the process of electron's relaxation led as side result to a decrease of the QE of abo ut one order of magnitude that for 532 nm resulted to be above 1%.

CONCLUSION

During last years new milestones in photoelectron beam properties have been achieved due to increase efforts on photocathode research: new world record average currents, sub-thermal transverse energy distribution. Photoemission properties of nanostructured photocathodes and multi-layered structures have been measured. Using tools typical of solid state physics new insight on the growth dynamics and electron transport have been obtained. Still there is a lack of complete characterization of many known and new materials and structures calling for more efforts from experimentalist to evaluate limits and advantages of different photocathodes.

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