

# IMPROVED LIFETIME OF A HIGH SPIN POLARIZATION SUPERLATTICE PHOTOCATHODE

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## Abstract

Highly spin polarized electron beams are required for the operation of a wide range of accelerators and instruments. The production of such electrons requires the use of Negative Electron Affinity (NEA) activated GaAs-based cathodes operated in photoelectron guns. Because of their extreme sensitivity to poor vacuum conditions the degradation of the photoemission process is so strong that NEA activated GaAs-based photocathodes can only survive in the extreme vacuums typical of DC gun. State-of-the-art on photocathode technology for spin polarized beam productions are summarized. Recent results on the use of robust NEA coating based on the Cs-Te and Cs-Sb leading to improved operational lifetime of a high spin polarization photocathode are reviewed.

## INTRODUCTION

The use of spin polarized electron source finds application in a wide range of electron accelerators. Along with the planned International Linear Collider [1] designed to collide spin polarized electrons and positrons at very large energies in the TeV scale, spin polarized electron beams are required for the next large accelerator facility for nuclear study that will be operated in the US: the Electron Ion Collider [2]. Aside from these very large scale facilities designed with the purpose to unravel new particles and the the fundamental laws of physics, the use of spin polarized electron beam finds also application in small scale accelerators for the study of magnetic properties of the materials like Spin Polarized Low Energy Electron Microscopy [3]. These large and small scale facilities require relatively modest amount of average current, not more that few hundred of micro Amperes that can be produced by state-of-the-art photoelectron sources. Other facilities like the Large Hadron electron Collider [4] are planning to operate with very high average electron beam currents, 20 mA, which are well beyond the current state-of-the-art. Also, the need of developing sources capable of providing high average beam power in the frame of spin polarized electron sources arises from the recent demonstration of an efficient transfer of spin polarization from electrons to positrons via a two-step process: In the first step polarized bremsstrahlung radiation is generated by a polarized electron beam in a high-Z target; and in the second step the same polarized bremsstrahlung radiation is used to produce polarized positrons by pair-production process in the same target [5]. Given the relative low efficiency of the process of production and capture of the positron beam ( $10^{-3}$ ) there

is the need of developing electron sources that can produce tens of mA of average spin polarized beam currents.

## STATE OF THE ART

When it comes to the production of spin polarized electrons beams in photoelectron guns there are currently not other option to GaAs-based photocathodes structures.

The discovery of spin polarized photoemission from GaAs was first reported in 1975 for bulk GaAs [6]. The electron beam spin polarization arises from the different probabilities associated to the optical transition from the energy degenerate heavy holes and light holes valence bands when circularly polarized light is used to excite photoelectrons. In bulk GaAs, large Quantum Efficiencies (QE) are possible (typically 0.1 electrons per number of incident photons) with Electron Spin Polarizations theoretically limited to a maximum value of 50%. In 1992, the use of a strained GaAs layer grown pseudomorphically on a relaxed GaAsP layer allowed breaking the energy degeneracy of the valence bands and an energy selective excitation of photoelectrons with a single spin polarization was demonstrated. Photoelectrons achieved ESP larger than 80%, but with typical QEs in the  $1e-3$  region [7]. The use of strained superlattices (SL) based on GaAs/GaAsP allowed to leverage the engineering of quantum wells structures to increase the separation between heavy hole and light hole bands [8] and achieve even larger spin polarization (up to 85%) and QE (just above 1%). Advanced SL design techniques made use of the strain compensation technique to decrease the defect density due to the accumulated strain in the SL. Structures are based on alternating layers with tensile and compressive strains so that on average the whole structure results unstrained [9]. Minimizing the defect density enabled further increase the spin polarization (up to 92%) with an achieved QE just above 2%. Even if SL strain compensated structures have been grown with a large number of quantum well (up to 90 pairs) the simultaneous maximization of the QE and ESP is hard to achieve. As QE increases because of the larger thickness of the SL medium, the ESP decreases because of depolarization mechanisms due to the interaction of the drifting electron with the lattice before their extraction into vacuum. A Distributed Bragg Reflector (DBR) grown underneath the SL lattice with a proper buffer layer separating the DBR and SL realize a Fabry Perot resonator that can effectively trap the light in a relatively narrow bandwidth inside the SL structure. Photons will be reflected back and forth between the DBR and the SL outer layers allowing to increase the light absorption and as consequence the QE. Such a structure demonstrated record performances achieving at the same wavelength (775 nm) an ESP of 84% and a QE of 6.4% [10].

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As the number of layers and materials increase the growth of such structure now becomes quite complicated (more than 50 layers and 5 different materials are required) affecting the rate of success in obtaining the design specification.

### *Negative Electron Affinity and Lifetime*

The electronic band gap of GaAs is about 1.4 eV and for reaching the photoemission threshold electrons must be excited using UV light. In order to extract electrons with photon energies close to the band gap in III-V semiconductor it is necessary to modify the surface electron affinity so that the vacuum level lies below the conduction band minimum. This condition is known as Negative Electron Affinity (NEA). This condition is achieved by exposing the clean GaAs surface to Cs vapors which result in the formation of strong electric dipole field near the surface that permit the extraction of electron that are relaxed to the bottom of the conduction band. Achieving the NEA condition usually require less than 1 mono-layer of Cs on the surface. This was confirmed by experimental data and numerical computations of work function variation as function of the Cs exposure as well as from advanced surface imaging technique [11, 12]. The strength of the surface dipole is enhanced by using alternating layers of Cs and O<sub>2</sub> or NF<sub>3</sub> gasses. Unfortunately, due to the extreme reactivity of these activating layers the vacuum requirements necessary to sustain the NEA condition are quite challenging: in order to have a sufficiently long lifetime to be of any practical use the GaAs surface activated to NEA are usually operated in vacuum levels below 10<sup>-11</sup> Torr. Such vacuum requirements have so far limited the operation of GaAs based photocathodes only on High Voltage DC electron guns. In these guns the vacuum vessel is usually very large to prevent the electric discharge between the cathode stalk which is held at high voltage and the surrounding UHV chamber walls at ground potential. The large volume of the vessel can host a large amount of Non Evaporable Getter pump providing large pumping speeds (many thousands of liter per second) and reaching vacuum levels of 10<sup>-12</sup> Torr or even better [13–15].

Dark lifetime measures the 1/e decay of the QE when cathodes are idling without electric field and charge extraction. In a typical DC gun designed to reach few 10<sup>-12</sup> Torr of base pressure dark lifetimes can easily reach few thousands of hours [16]. In such extreme vacuum and in the absence of laser illumination and electric field allowing for electron extraction, the only mechanisms responsible for the degradation of the efficiency is the chemical poisoning of the activation layer in response to the exposure of gas molecules in the residual gas: even in the extreme vacuum small quantities of oxygen containing species are still present and for some of them, like O<sub>2</sub> and CO<sub>2</sub>, only a fraction of a monolayer is sufficient to produce dramatic reduction of photoemission [16].

When the photocathode is used to generate electrons a better metric to define the lifetime is perhaps the charge lifetime which measure the 1/e decay of the quantum efficiency as function of the extracted integrated charge. In this case

the QE degradation is affected not only by the chemical poisoning, but also by the chemical desorption of alkali metals induced by the thermal laser heating of the photocathode surface [17] and by the back bombardment of ions accelerated by the field existing in the anode-cathode gap and striking at the photocathode surface [18]. The number of experimental parameters influencing the scaling laws associated to these last two degrading processes and the way they are entwined complicates the scenario and it is often difficult comparing results obtained with different electron guns or distinct experimental conditions. Indeed, ions in the cathode anode gap are usually produced by ionization of the residual gas molecules by electron impact from the primary electron beam and their number will be proportional to the gas density in that region. On the other hand the gas density may also be influenced by electron induced desorption from stray energetic electron hitting the surrounding surfaces which are difficult to quantify and predict.

At average beam currents levels of 1 mA and 4 mA charge lifetimes of 200 C and 80 C have been reported [19, 20] corresponding to a 1/e lifetime during operation of about 2.5 days and 5 hours respectively. Taking the most favorable case (200 C of charge lifetime) and scaling it for an average current of 20 mA (like the one required by the ERL proposed in LHeC) will result in a operating 1/e lifetime of less than 3 hours. These estimates seems to be quite well aligned also with the operational lifetime measured at the DC gun of Cornell University where GaAs was operated at record average currents up to 50 mA showed 1/e QE lifetimes of about 1 hour [21].

## **ROBUST NEA ACTIVATING COATING**

Given the extreme sensitivity of GaAs NEA surfaces to vacuum requirement is it not surprising that even before the spin polarized photoemission from GaAs was firstly observed, there was interest in developing robust methods to generate the NEA on this family of semiconductors. The first report showing that a Cs-Sb layer can produce NEA on GaAs [22] was reported in 1969 along with the first speculation on the possible formation of a p-n hetero-junction between the two materials [23]. Hetero-structured photocathodes based on bulk GaAs activated with Cs and Sb to NEA were again reported in 1993 showing that an increase of the longevity was achieved with such activation [24]. In 1996, another report confirmed the increase of the lifetime up to a factor 15 and also indicated that different composition of alkali antimonides surface layer can be used to provide NEA on GaAs [25]. Several years later the measure of the energy shift in the electron energy distribution from GaAs coated with a very thin layer of Cs-Te the NEA indicated a lowering of the surface workfunction of about 3.1 eV, sufficient for the formation of Negative Electron Affinity [26]. Further experiments aimed at optimizing this activation procedure and the thickness of the coating confirmed the NEA achievement as it was deduced from spectral response measurement showing electron emission at photon energies equal to the band

gap [27]. This was a quite relevant result: the  $\text{Cs}_2\text{Te}$  is a well known material in the photocathode community because it is a robust semiconductor photocathode materials that is widely operated in RF guns. Facilities like FLASH [28], the European X-FEL [29], LCLS-II [30], AWA [31] and many other labs are using  $\text{Cs}_2\text{Te}$  photocathodes in their photoinjectors. The demonstration of  $\text{Cs}_2\text{Te}$  based activation could potentially open the path to the operation of GaAs based photocathodes in RF and SRF guns, enabling the long term storage of photocathodes and even their transport into vacuum suitcases. There was the need to answer to a last important question: will the Cs-Te activation layer preserve the electron spin polarization during the transport and emission into vacuum? It is important to stress that while we have now some answers related to this question, the results so far reported in literature and related to the use of robust coatings for generating NEA on GaAs based photocathodes have been obtained at very low extraction fields, with electron energies of only few tens of eV and not yet a beam energies typical of modern electron guns. For this reason, a direct comparison with the lifetime performances obtained in a state-of-the-art electron guns are somehow difficult. The relevance of the ion back bombardment process in decreasing the QE is largely influenced by the electron impact ionization cross sections, which is 3 order of magnitude larger at the energy where the upcoming result are obtained than the ones typical of modern photoinjector. In addition the vacuum levels at which the surface science studies have been carried out were in the  $10^{-11}$  Torr, one order of magnitude larger than the typical vacuum achieved in HV DC gun indicating a larger residual gas density. Finally, the kinetic energies of the ions accelerated towards the cathode is low enough (few tens of eV at most) that any damage induced by their impact would involve only the very surface layers which are the most critical for the NEA condition.

### The Cesium Telluride Activating Coating

A possible band diagram of an hetero-junction based on p-type GaAs and  $\text{Cs}_2\text{Te}$  is shown in Fig. 1. Achieving a band alignment like the one showed in the Fig. 1, that sees the vacuum level on the  $\text{Cs}_2\text{Te}$  vacuum interface at the same level of the conduction band minimum of the GaAs, requires an n-type doped layer of  $\text{Cs}_2\text{Te}$ . Such doping could be possibly achieved by incorporating excess Cs in the film during the growth process. In practice, the cesium telluride layers are grown using the photocurrent extracted during the growth as feedback for the whole process and a fine control of the doping density has never really put in place.

Details of the growth procedure used by Cornell University group can be found in ref [32]. The estimated total thickness for the cesium telluride was in the range of 2-2.5 nm. After the growth of the cesium telluride layer the spectral response showed photoemission at photon energies equal to the band gap confirming that NEA condition was achieved. A QE of about 1% was obtained at the wavelength of 780 nm which is where the largest spin polarization in bulk GaAs is achieved.

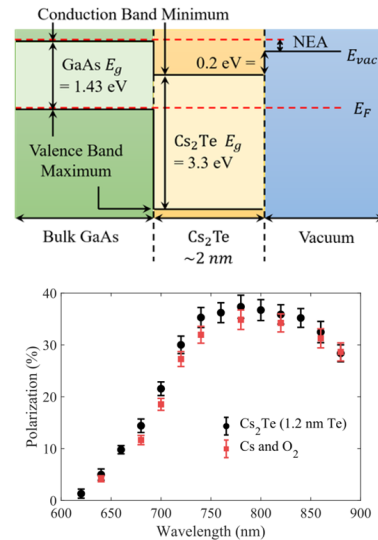


Figure 1: Scheme of the band diagram of the hetero-structure GaAs/ $\text{Cs}_2\text{Te}$  and result from the spin polarization measurements. Reproduced from [32].

A BNL group was able to further improve the quantum efficiency by adding to the simple cesium telluride growth recipe the use of Cs and oxygen [33]. With the adoption also of a tailored GaAs surface heat cleaning procedure (580 C for about 2 hours) aimed at minimizing the surface roughness and still obtaining a surface free of most contaminants [34], they were able to increase the efficiency at 780 nm up to about 4%. The lifetime of the GaAs samples activated with Cs-Te was characterized by and compared with samples activated with the standard Cs-O method. Detailed result were obtained at Cornell University indicating that an increase of the lifetime of a factor 5 can be achieved using the activation with cesium and tellurium if compared to standard activation with Cs and oxygen [32]. Similar improvement on the lifetime were observed also from the BNL group which reported a factor 6 increase in the lifetime [35]. The rejuvenation of the photocathodes activated with Cs and Te can be achieved with an additional exposure to Cs vapors but the lifetime after this procedure resulted to be comparable to the one characteristics of a Cs and oxygen activation [32]. The most important observation was that the electron spin polarization was not affected by the cesium telluride coating and no difference were observed in the measurement for the same GaAs specimen that was activated to NEA first using Cs and oxygen and later using Cs and Te [32].

### The Cesium Antimonide Activating Coating

Another activating layer that was studied in quite detail make uses of Cs, Sb and oxygen. Basing the hetero-junction model on the smaller band gap of the  $\text{Cs}_3\text{Sb}$  it is expected that the NEA can be achieved with an activating layer with no net doping removing the need of incorporating additional Cs to increase the n-doping and causing the required shift of the Fermi level in the  $\text{Cs}_3\text{Sb}$  towards the bottom of the conduction band. Different growth procedures were studied



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at Cornell University to find the optimal configuration that could simultaneously maximize the QE and the lifetime. The oxygen was used in the attempt of forming a Cs-O dipole layer that would eventually segregate at the interface between the GaAs and the Cs-Sb layer and also as an additional termination layer at the vacuum interface to further lower the electron affinity at the surface [36]. All the different procedures used the exact same amount of Sb for the activation to reach the negative electron affinity conditions and resulted in an electron spin polarization that was unaffected by the particular method used for the activation. Eventually, from the combination of QE and lifetime measurements it was observed that the activation procedure that used a continuous exposure to oxygen gas during the growth of the Cs and Sb layers allowed to achieve simultaneously the best performances in terms of efficiency and robustness [36]. An-

ures to Sb vapors were used. The methods for the activation was chosen to be the one that used Cs and oxygen during the entire growth of the activating layer and that gave the best results in the previously reported set of experiment [37]. The results (see Fig. 2) indicated that as the Sb thickness is increased, leading to an overall thicker activating layer, the NEA condition is always achieved but the efficiency of the photoemission process decreases (about one order of magnitude lowering in the QE was measured as the Sb thickness increased from 0 to 0.5 nm). On the other hand, the robustness of the photocathode seems to increase and longer lifetimes are measured as the thickness of the initial Sb layer is increased: up to a factor 10 increase in dark lifetime and up to 60 increase of charge lifetimes have been estimated from QE vs. time measurement carried out with duty cycles of 2% and 100%. Finally, as the Sb layer thickness is increased to largest value the maximum electron spin polarization achieved by the samples is seen to progressively decrease from the initial value of 38% for the Cs and oxygen layer to 25% [37]. A simplified model taking into consideration the estimated width increase of a triangular surface barrier with the Sb thickness seems to qualitatively be able to reproduce the main feature of the experimental observations. On the overall, the set of experimental data allow generating a picture of the trade-offs that seems to be required for increasing the lifetime using these robust NEA coatings: at the photon wavelength of 780 nm, corresponding to the photon energy where the electron spin polarization in GaAs is maximal, the QE decreases by a rough factor 2 to 3 but the spin polarization is still unaffected if an initial Sb layer of .13 nm is used to grow the surface layer. With this Sb thickness about one order of magnitude longer charge and dark lifetimes are achieved and the total activating layer thickness is expected to be slightly larger than 1 nm. By accepting more losses in QE and electron spin polarization further gains in lifetime can be achieved using larger amounts of Sb [37]. All these results were achieved using bulk GaAs and it is important to demonstrate the applicability of these methods to a photocathode structure consisting of a SL designed to provide the highest electron spin polarization. TO this scope the Jlab injector group provided Cornell University group with a sample from a SL GaAs/GaAsP wafer. Photocathodes from the same wafer were extensively used in the past years in CEBAF photoinjector delivering electron beam with spin polarization of about 90%. The sample was activated using the Cs-Sb-oxygen recipe developed using bulk GaAs samples and with the Sb layer tuned to yield no polarization losses. The comparative results with a standard Cs-O activation are well aligned with the expectations (see Fig. 3): QE is decreased by factor 3, electron spin polarization remain unchanged, the operational lifetime increases by a factor 7 [36].

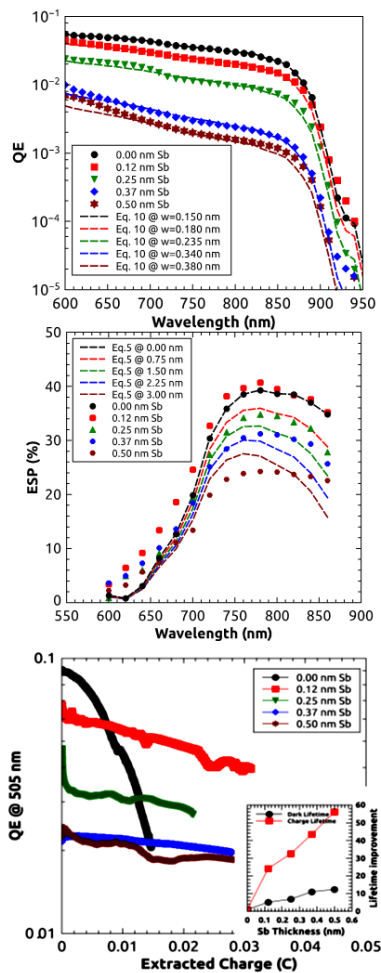


Figure 2: From top to bottom: Spectral response, electron spin polarization and lifetime of GaAs samples activated to NEA with different thicknesses of the Cs-Sb-O layer. Reproduced from [37].

other set of experiments focused the attention on the role played by the thickness of the activating layer in the electron emission performances (QE , ESP, lifetime). To generate different thicknesses of the activating layer increased expo-

## CONCLUSION

The increase of the lifetime of GaAs based photocathodes for spin polarized production can be achieved using robust

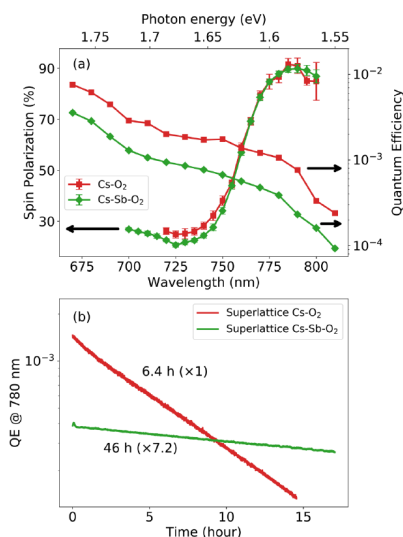


Figure 3: From top to bottom: Spectral response, electron spin polarization and lifetime of a superlattice GaAs/GaAsP sample activated to NEA with Cs-Sb-O layer. Reproduced from [36].

activating layers based on Cs-Te-O or Cs-Sb-O. The losses in QE and electron spin polarization arising from the electron scattering in the activation layer should be taken into account as unavoidable trade-offs to increase the lifetime. A simultaneous use of Distributed Bragg Reflector (to maximize the light absorption in the SL) and of strain compensated SL (to maximize the QE and ESP generated in the SL) can be used to gain margins for these trade-offs. It is of utmost importance validating the results so far obtained in a surface science experimental setting also in state-of-the-art electron gun and gather data on the performance of these photocathodes in real operational condition. The SL and SL-DBR are difficult to obtain: they cannot be simply purchased off the shelf and they must be grown as custom design. Maintaining and operating the required equipment to perform the growth of these materials is a non trivial task in terms of investment and required skills. It is important for the community to seek and facilitate collaboration with other lab and facilities that can provide the required expertise. Cornell University and Jlab have teamed up to perform tests of the robust activating layers at the UITF facility at Jlab. The experiments, delayed because of the COVID-19 pandemic, aim at upgrading the existing beamline and perform the robust activation of GaAs-based photocathodes and measure QE, electron spin polarization and lifetime at the highest possible current (few mA). While the losses in QE due to the robust coating can be compensated by using increased level of laser power and yet achieve the design average current or bunch charge, not much can be done to restore the spin polarization lost in the robust activating layer. A possible channel that can be leveraged to increase the initial spin polarization and compensate for some of the losses is the operation of the photocathode at cryogenic temperatures: this will strongly mitigate the depolarization processes inside the bulk or SL photocath-

ode material possibly yielding ESP closer to 100% [38]. The development of electron guns capable to maintain the photocathode at cryogenic temperature [39–41] opens the possibility of leveraging the cryocooling of the photocathode to mitigate the spin relaxation rates of electrons inside the material. Again, dedicated experiments should be performed to understand to what extent the performance can be quantitatively improved.

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