ISBN: 978-3-95450-247-9 ISSN: 2673-5490 doi: 10.18429/JACoW-IPAC2024-MOPR79

# DEMONSTRATION OF ENHANCED QUANTUM EFFICIENCY FROM OPTICAL INTERFERENCE IN ALKALI ANTIMONIDE PHOTOCATHODES

C. Pennington*, E. Echeverria, Jared Maxson, Cornell University, Ithaca (NY), USA
M. Gaowei, K. P. Mondal, K. Evans-Lutterodt, Brookhaven National Laboratory, Upton (NY) , USA
J. Smedley, SLAC, Menlo Park (CA), USA
P. Bhattacharyya, S. Karkare, Arizona State University, Tempe (AZ), USA
A. Galdi, University of Salerno, Fisciano (SA), Italy

*Abstract*

We present measurements of enhanced quantum efficiency (QE) in thin film photocathodes due to optical interference in the cathode-substrate multilayer. Modulations in the quantum efficiency of $Cs_3Sb$ films grown on multilayer 3C-SiC substrates are observed over a range of visible wavelengths, and are shown to increase the QE by more than a factor of two at certain wavelengths. We derive a model to describe the modulations in QE based on a three step photoemission process incorporating cases of both constant density of states and density functional theory (DFT) derived density of states, which is shown to be in good agreement with the measurements. Predictions from the model show that the QE can be enhanced by more than a factor of four by optimizing the cathode and substrate layer thicknesses. We also find that by optimizing layer thicknesses of the cathode-substrate system in the calculation, optical interference can enhance the QE beyond optically dense films. Advantages of this interference effect for electron accelerator sources are discussed.

## INTRODUCTION

Enhancing the quantum efficiency (QE) and performance of photoemission sources is an active area of development for electron accelerators to produce high brightness beams [1]. The effect of photocathode material properties on QE performance, such as chemical composition, surface roughness, and crystallinity, are actively studied [2-4]. Previous works aim to enhance QE via optical effects such as waveguide structures [5], while theoretical analyses demonstrate that optical interference can enhance the QE by over an order of magnitude for widely studied photoemitting materials and substrates [6]. Significant QE enhancement has also been demonstrated from photocathodes grown on distributed Bragg reflectors (DBRs) [7, 8].

In this article, we present measurements of quantum efficiency enhancement in alkali antimonide photocathodes due to optical interference in the cathode-substrate multilayer. Thin films of alkali antimonides were grown on a multilayer substrate, effectively a DBR, consisting of a layer 3C-SiC that was epitaxially grown on a thicker layer of silicon. Increasing the QE using optical interference aims to complement the work on optimizing material and morphological properties to improve alkali antimonide photocathode performance [3, 9].

* cap354@cornell.edu

## THIN FILM SYNTHESIS

Thin films of $Cs_3Sb$ were grown on 3C-SiC(001) substrates Fig.1 in a molecular beam epitaxy (MBE) chamber at the Photocathode Epitaxy and Beam Experiments (PHOEBE) Laboratory at Cornell University. The substrates were degassed at temperatures between 450°C-500°C for 30 min until a clear RHEED pattern of the substrate surface was visible. Vacuum levels were maintained near $1\times10^{-9}$ Torr during film deposition. The growth method was similar to the one described in [3], using a two-temperature profile from 50°C to 90°C.

The films were moved to an adjacent UHV sample chamber ($P\approx1\times10^{-10}$ Torr) for spectral response measurements. An Oriel Apex Monochromator light source was used to generate the visible spectrum, as well as a model 843-R Newport optical powermeter, and a SRS 8340 lock-in amplifier to measure QE. Photocurrent was collected using biased metallic coil (120 V) placed 5 cm from the photocathode sample.

The CsSb films were grown on 3C-SiC(001) substrates at the National Synchrotron Light Source II (NSLS-II) of Brookhaven National Laboratory (BNL) at beamline 4-ID ISR. The photocathode growth chamber and experimental apparatus are described in detail in [9, 10].

## RESULTS AND DISCUSSION

QE oscillations were observed in the spectral response of a thin layer of $Cs_3Sb$ on the 1.3 $\mu$m thick 3C-SiC substrate shown in Fi.2(a). The QE modulates by up to a factor of two, within a 30 nm period. We observe this effect again using a thinner layer, 0.3 $\mu$m thick, of 3C-SiC as shown in Fig.2(b). The dependence of the oscillation period on thickness indicates thin film interference.

We present a model to further understand the variables that affect the depth of modulation and periodicity of the QE from optical interference. We consider two cases in the model: the first is a photocathode material with constant density of states (DOS), and the second is with a DOS calculated from density functional theory DFT.

ISBN: 978-3-95450-247-9 ISSN: 2673-5490 doi: 10.18429/JACoW-IPAC2024-MOPR79

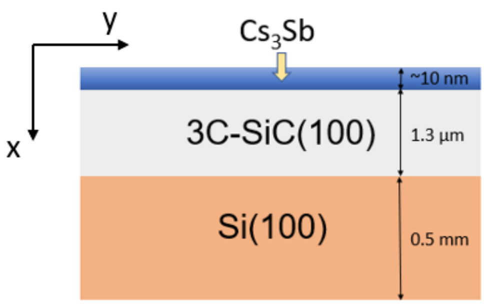


Figure 1: Schematic of a multilayer substrate used in this experiment. 3C-SiC(001) is grown epitaxially on Si(001). Two 3C-SiC layer thicknesses were used in this experiment: 0.3 $\mu$m and 1.3 $\mu$m. The photocathode thicknesses are estimated to be between 5 nm and 20 nm depending on the sample.

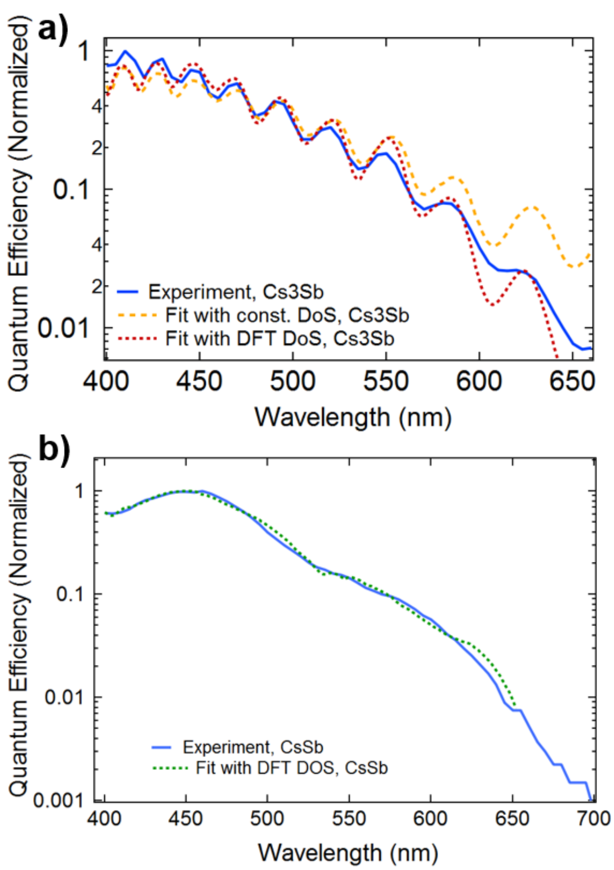


Figure 2: (a) Calculated QE vs measured QE of $Cs_3Sb$ grown on 3C-SiC for SiC layer thicknesses of (a) 1.3$\mu$m, with fits including a constant DOS assumption and a density functional theory calculated DOS and (b) A 0.3$\mu$m.

For the case of constant DOS, the QE is proportional to the square of the excess energy, as shown in [11], before considering optical interference. With optical interference, the absorption in the photocathode layer varies with depth in a nontrivial way. We define an enhancement factor, $\mathcal{F}_e(\hbar\nu)$, to account for this variation, where $h$ is Planck's constant and $\nu = \frac{c}{\lambda}$ where $\lambda$ is the laser wavelength and $c$ is the speed of light. This gives us the relation:

$$QE \propto (h\nu - \phi)^2 \mathcal{F}_e(h\nu) \quad (1)$$

where $\phi$ is the workfunction of the material and $h\nu - \phi$ is the excess energy.

We derive the enhancement factor by calculating the total electric field in the photocathode film:

$$E_y = E_o e^{i\omega t}\left(c_f e^{i\tilde{n}_{\mathrm{PC}} kx} + c_b e^{-i\tilde{n}_{\mathrm{PC}} kx}\right) \quad (2)$$

where $c_f$ and $c_b$ are the coefficients of the forward and backward travelling waves and $n_{\mathrm{PC}}$ is the complex index refraction of the material. We solve the for coefficients $c_f$ and $c_b$ by applying boundary conditions for the electric field in the multilayer, then calculate the net power density flow from the Poynting vector $\langle S_x \rangle = \frac{1}{2} E_y \times H_z^*$, where $H_z^*$ is the complex conjugate of the magnetic field in the material which can be derived from $H = (-1/\mu) \int \nabla \times E dt$. The power density is then defined as $P_a = \frac{\partial}{\partial x} \mathrm{Re} \langle S_x \rangle$, and a power absorption profile $a(x)$ in the photocathode is defined as: $a(x) = P_a / P_{\mathrm{in}}$, where the input power at the surface, $P_{\mathrm{in}}$, is calculated from:

$$P_{\mathrm{in}} = -\left(\frac{k n_{\mathrm{vac}}}{2\omega\mu_o\mu_V}\right) |E_o|^2 . \quad (3)$$

After calculating the absorption profile $a(x)$, we assume the probability of an electron escaping from the material decays exponentially with thickness, and calculate the enhancement factor:

$$\mathcal{F}_e = \int_0^l a(x) e^{-(x/\lambda_{esc})} dx \quad (4)$$

where $l$ is the thickness of the photocathode layer, and $\lambda_{esc}$ is defined as the electron escape depth of the material. The QE is then modeled for the constant DOS case as:

$$QE(h\nu) = N(h\nu - \phi)^2 \int_0^l a(x) e^{-(x/\lambda_{esc})} dx. \quad (5)$$

To make the model more robust at predicting QE enhancement, we include a detailed DOS obtained by density functional theory (DFT) in the calculation. To account for variable DOS in the model, we again follow the derivation of the QE from Saha in [11] to express the quantum efficiency shown in Eq. (6).

We then input data on the optical properties for Si and 3C-SiC obtained from [12,13] and the complex refractive indices for $Cs_3Sb$ obtained from DFT calculations to calculate the QE in Eq.(5). The thickness of the SiC layer, thickness of the

ISBN: 978-3-95450-247-9 ISSN: 2673-5490 doi: 10.18429/JACoW-IPAC2024-MOPR79

$$QE(h\nu) = N\frac{\int_{-\infty}^{\infty} D(E)f(E)D(E+h\nu)[1-(f(E+h\nu)]EdE}{\int_{-\infty}^{\infty} D(E)f(E)D(E+h\nu)[1-(f(E+h\nu)]dE}\int_0^h a(x)e^{-(x/\lambda_{esc})}dx \quad (6)$$

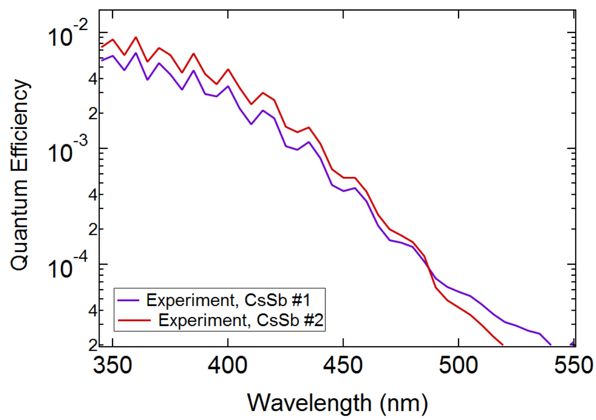


Figure 3: (a) Measured QE of $Cs_1Sb_1$ on a 1.3$\mu$m thick SiC layer. A thinner $Cs_1Sb_1$ cathode (blue curve) is compared to a thicker cathode (red curve).

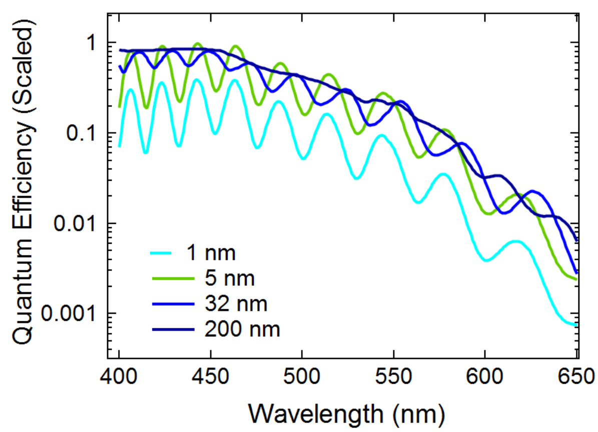


Figure 4: Calculated QE for $Cs_3Sb$ cathodes with different thicknesses. The substrate used in the model is 1.3 $\mu$m thick SiC on 0.5 mm thick Si.

$Cs_3Sb$ layer, the electron escape depth $\lambda_{exc}$, and the vacuum level energy, $E_{vac}$ are free parameters used to fit to the data. The resulting fit is shown with the measurements in Fig. 2 for both cases of constant DOS and DTF calculated DOS.

Optical interference was also observed for the growth of $Cs_1Sb_1$ photocathodes at BNL as shown in Fig.3. To date, the refractive index has not been directly measured on this stoichiometry of cesium antimonide. We plan to apply the model to $Cs_1Sb_1$ and other materials after obtaining DFT calculations or measurements of the refractive indices.

In Fig.4, we test the model by varying thicknesses of the $Cs_3Sb$ layer while keeping the SiC layer fixed at 1.3 $\mu$m and holding all other parameters constant. We find that the QE modulates by a factor greater than four in a 5 nm thick Cs3Sb cathode. The optical interference from the 5 nm thick film will enhance the QE beyond that of an optically dense, 200 nm thick cathode by a factor of 1.33 at 515 nm, and by large fractions of the QE at several constructively interfering wavelengths. The implication of this calculation is that optical interference allows thinner cathodes to provide larger QE at desired visible wavelengths.

Dampening of the QE modulation depth for thicker films is largely due to the absorption of light in the cathode. The modulations would disappear completely in an optically dense cathode, which has thickness greater than the absorption depth of the light. Surface roughness and more detailed scattering processes could also affect the QE, however modeling these effects are outside the scope of this article.

## CONCLUSION

Enhanced quantum efficiency be achieved by exploiting optical interference effects in multilayer photocathode-substrate systems. We present measurements of QE modulations in $Cs_3Sb$ cathodes grown on 3C-SiC(001) substrates that form this multilayer system, and show that the QE more than doubles over a wavelength range of approximately 30 nm. Our model of the optical interference effect is shown to be in good agreement with the measurements, confirming the effect is thin film interference. Predictions from the model show that the QE can be enhanced by more than a factor of four using our conditions, and that the QE can exceed that of an optically dense cathode by a factor greater than 1.33 by optimizing layer thicknesses. The modulation depth and period of the QE oscillations can be controlled by tuning the substrate and photocathode layer thicknesses, allowing the QE to be enhanced at desired wavelengths for photocathode-based accelerators.

## ACKNOWLEDGMENTS

The authors thank M. Gordon, M. Kaemingk, and A. Bartnik for helpful discussions on this work and for checking derivations. The authors thank C. Parzyck and T. Arias for providing useful data from DFT calculations. [1]

## REFERENCES

[1] P. Musumeci, *et. al.*, "Advances in bright electron sources", *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 907, pp. 209-220, Nov. 2018. doi:10.1016/j.nima.2018.03.019

[1] This work was supported by the National Science Foundation award No. PHY-1549132. This work was supported in part by DOE grant DE-AC02-76SF00515. Work supported by Brookhaven Science Associates, LLC under Con- tract No. DE-SC0012704, DE-SC0013190 with the U.S. Department of Energy. The use of National Synchrotron Light Source II at Brookhaven National Laboratory is supported by U.S. Department of Energy Office of Science under Contract No. DE-AC02-98CH10886.

ISBN: 978-3-95450-247-9 ISSN: 2673-5490 doi: 10.18429/JACoW-IPAC2024-MOPR79

[2] Jun Feng, *et. al.*, "Near atomically smooth alkali antimonide photocathode thin films", *Journal of Applied Physics*, vol. 121, no. 4, p. 044904, Jan. 2017. doi:10.1063/1.4974363

[3] C.T. Parzyck *et al.*, "Single-Crystal Alkali Antimonide Photocathodes: High Efficiency in the Ultrathin Limit", *Physical review letters*, vol. 128, p.114801, Dec. 2021.

[4] D. Dimitrov *et al.*, "Modeling optical interference effects for optimization of electron emission properties from thin film semiconductor photocathodes", in *Proc. IPAC'23*, Venice, Italy, May 2023, pp. 1378–1381. doi:10.18429/JACoW-IPAC2023-TUPA012

[5] P D Townsend *et al.*, "Designs for waveguide and structured photocathodes with high quantum efficiency". *Journal of Physics D: Applied Physics*, vol. 39, no. 8, p. 1525, Mar. 2006. doi:10.1088/0022-3727/39/8/012

[6] A. Alexander *et. al.*, "Enhanced photocathode performance through optimization of film thickness and substrate", *Journal of Vacuum Science and Technology B*, vol. 35, p. 022202, Mar. 2017. doi:10.1116/1.4976527

[7] Jyoti Biswas *et. al.*, "Record quantum efficiency from strain compensated superlattice GaAs/GaAsP photocathode for spin polarized electron source", *AIP Advances 1*, vol. 13, no. 8, p. 085106, Aug. 2023. doi:10.1063/5.0159183

[8] Wei Liu *et. al.*, "Record-level quantum efficiency from a high polarization strained GaAs/GaAsP superlattice photocathode with distributed Bragg reflector", *Appl. Phys. Lett*, vol. 109, no. 25, p. 252104, Dec. 2016. doi:10.1063/1.4972180

[9] C.A. Pennington *et al.*, "A structural analysis of ordered Cs3Sb films grown on lattice-mamt graphene and silicon carbide substrates", unpublished.

[10] W.G. Stam *et al.*, "Growth of ultra-flat ultra-thin alkali antimonide photocathode films", unpublished.

[11] Pallavi Saha *et al.*, "Theory of photoemission from cathodes with disordered surfaces", *J. Appl. Phys.*, col. 133, no.5 , p. 053102, Feb. 2023. doi:10.1063/5.0135629

[12] Stuart M. Johnson, "Optical absorption and photoemission in semitransparent and opaque Cs3Sb photocathodes", *Appl. Opt.*,vol. 32, pp. 2262-2265, 1993.

[13] M. N. Polyanskiy. "Refractiveindex.info database of optical constants", *Sci. Data* , vol. 11, p. 94, Jan. 2024. doi:10.1038/s41597-023-02898-2