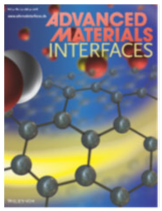
Free-standing bialkali photocathodes using atomically thin substrates

H.Yamaguchi[1]*, F.Liu[1], J.DeFazio[2], M.Gaowei[3], C.W.Narvaez Villarrubia[1], J.Xie[4], J.Sinsheimer[3], D.Strom[5], V.Pavlenko[1], K.L.Jensen[6], J.Smedley[3], N.A.Moody[1]#

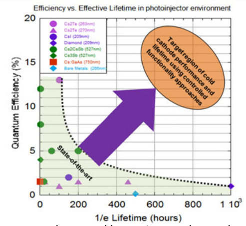
[1]Los Alamos National Laboratory, NM 87545, USA, [2]Photonis Scientific Inc., PA 17601, USA, [3]Brookhaven National Laboratory, NY 11973, USA, [4]Argonne National Laboratory, IL 60439, USA, [5]Max Planck Institute for Physics, Germany, [6]Naval Research Laboratory, Washington DC 20375, USA

Abstract



Enhancing the lifetime of accelerator technology-relevant photocathodes while maintaining their high quantum efficiency has been a decadal problem. Here, we successfully deposited Bialkali antimonide photocathodes on free-standing atomically thin substrates of graphene. This advances an ultimate goal of enhancing the lifetime of photocathodes without sacrificing the high quantum efficiency by using an atomically thin protection layer.

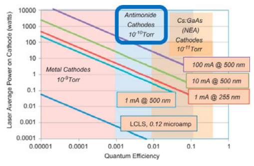
Driving Science Question



The required advances in cold cathode performance requires that radical improvements in lifetime and efficiency be achieved simultaneously. Previous work has succeeded in delivering increases in one, but at considerable expense of the other due to the competing physical processes underlying traditional approaches to cathode design and optimization. In pursuit of this goal, the unique approach started here aims to decouple the competing mechanisms so that both high efficiency and long lifetime can be achieved via integration of atomically thin two dimensional (2D) nanomaterials with high-performing existing photocathode technologies.

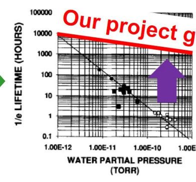
Background

High quantum efficiency (QE)



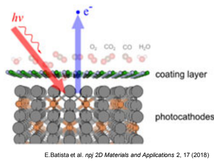
Semiconductor antimonide cathodes performance

...but susceptible



Cathodes lifetime *versus* water pressure

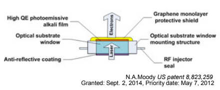
Atomically thin protection!!



Our project concept

Our previous work timeline

2012 Patent filed



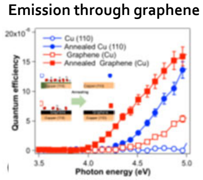
2014 Patent granted

2014 Project started

Three-year internal project was funded at LANL to form a team and start the project.

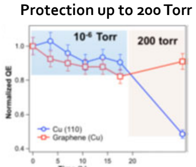
2017 First demonstration on metal

Emission through graphene



Graphene, atomically thin sheet of carbon, was directly grown on copper metal cathode as a protection layer. We demonstrated for the first time that photoemission is not hindered by the presence of graphene.

Protection up to 200 Torr



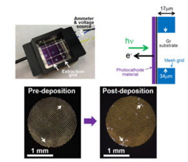
Furthermore, we found that graphene protects metal cathodes from residual gases in the environment. Graphene was able to protect metal cathodes to the pressure of 200 Torr, which could be a significant advancement toward cathode transfers in a near ambient condition.

F.Liu et al. Appl. Phys. Lett. 110, 041607 (2017)

Progress and Results

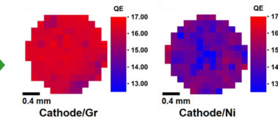
2018

K_2CsSb photocathode deposition on graphene (Photonis Scientific Inc.)

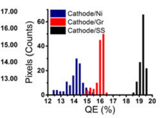


Metal mesh grid supports were used to prepare free-standing graphene substrates. Note minimal voids even after the photocathode deposition.

Overall QE maps



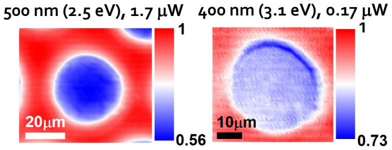
Statistics



~17 % QE achieved from K_2CsSb on graphene substrates. This was higher than that on nickel substrate. Spot size is 0.2 mm.

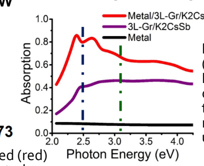
High spatial resolution QE maps

500 nm (2.5 eV), 1.7 μW — 400 nm (3.1 eV), 0.17 μW



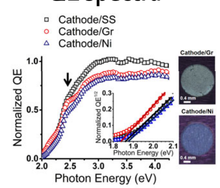
The maps clearly resolve freely suspended (blue) and supported (red) regions. QE ~10 % for the suspended and ~ 17 % on the supported regions were achieved for 2.5 eV photons. Spot size is 350 nm.

Calculated optical absorption spectra



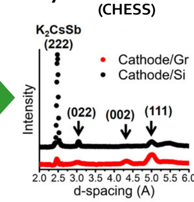
NKD Gen software (University of Barcelona) based on conventional transfer matrix methodology was used.

QE spectra



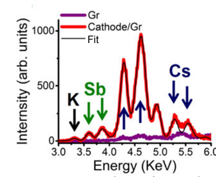
Signature feature for K_2CsSb was observed at ~2.4 eV (black arrow). Extracted work functions were all consistent with a value for K_2CsSb, which were 1.84, 1.86, and 1.88 eV for graphene, SS, and Ni substrate regions, respectively as shown in inset.

Synchrotron XRD (CHESS)



	d-spacing (Å)	Width (Å)	(HKL)	Theory (Å)
Cathode/Si	2.48	0.05	(222)	2.49
	3.05	0.08	(022)	3.05
	5.00	0.16	(111)	4.98
Cathode/Gr	2.47	0.09	(222)	2.49
	3.01	0.16	(022)	3.05
	4.31	-	(002)	4.31
	5.00	0.23	(111)	4.98

Synchrotron XRF (CHESS)



XRD spectra indicated K_2CsSb crystal growth on graphene. Quantitative analysis of XRF spectra resulted in elemental composition of $K_{1.85}Cs_{1.08}Sb$.

H.Yamaguchi et al. Advanced Materials Interfaces 5, 1800249 (2018)

Concluding Summary

In summary, bialkali antimonide photocathodes were successfully grown on freely suspended graphene substrates of few-monolayer thickness. The large-area uniformity, quantum efficiencies, specific spectral response features, and comparisons to experimental reference standards and calculation results all indicate a high compatibility of the photocathodes with thin graphene layers. Additional material characterization using XRD and XRF directly revealed grains of K2CsSb in such photocathodes when deposited on graphene, further supporting this compatibility. Variations in the number of graphene layers, their stacking methods, and additional pre-deposition annealing variations had small effects on the QE magnitudes of the resultant photocathodes, yet highlighted areas for measurable improvements by minimizing defects in the graphene films. The results demonstrate that high QEs can be achieved for K2CsSb on thin graphene substrates that are as thin as three atomic layers. This is a promising step toward fabrication of photocathodes with enhanced lifetimes via atomically thin protection layers as well as photocathodes on optically transparent yet electrically conductive substrates.

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*Primary: hyamaguchi@lanl.gov; #Secondary: nmoody@lanl.gov