Long Lifetime Spin-polarized Electron Sources: high current performance of alternative GaAs activation materials and novel spin-polarized sources via epitaxial growth

Matthew Andorf

Ivan Bazarov (PI)

Outline

- Motivation
- Cornell Photocathode Laboratory, HERACLES, PHOEBE
- Gun tests of alternative NEA activation coatings for GaAs
  - New growth chamber status
  - Updated laser system for HERACLES
- Alkaline-antimonide photocathodes as a spin polarized source?
  - DFT computations

HERACLES Team

Acknowledgements to:
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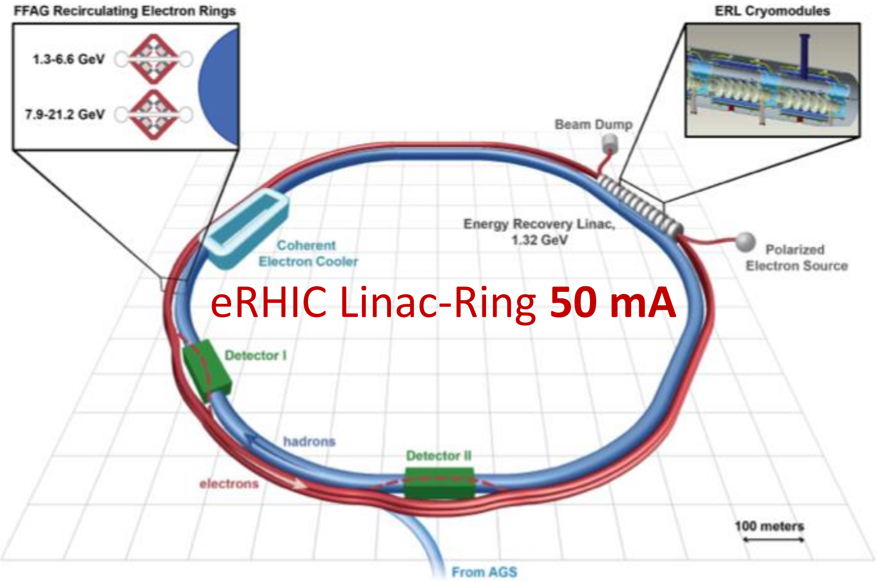
- Ivan Bazarov
- Jared Maxson
- Adam Bartnik
- Alice Galdi (now @U. of Salerno)
- **Sam Levenson (graduate student)**
- **Mark Reamon (undergrad)**

HERACLES

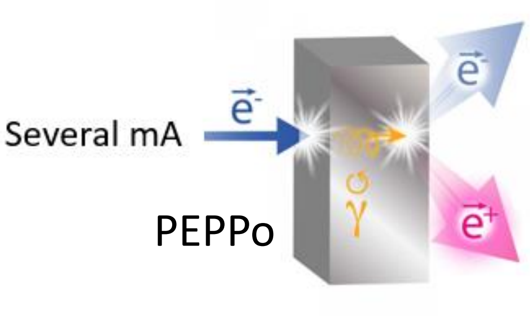
HIGH ELECTRON
AVERAGE CURRENT FOR
LIFETIME EXPERIMENTS

High current and spin polarization



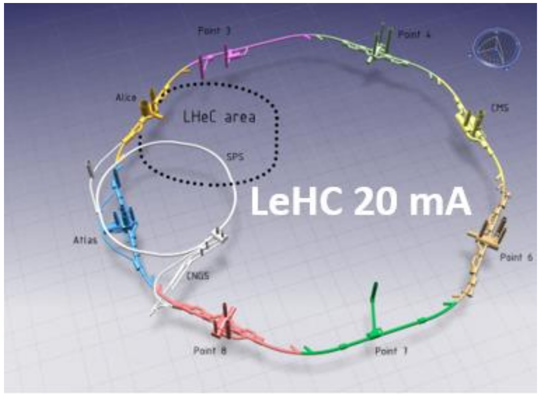
eRHIC Linac-Ring **50 mA**

- Many future facilities want spin-polarized electrons at high average currents
- Only candidate cathode for the job: GaAs!
- **Charge lifetime**: The amount of current extracted before the QE degrades by 1/e
  - 1000 C is state of the art→6 hours at 50 ma!

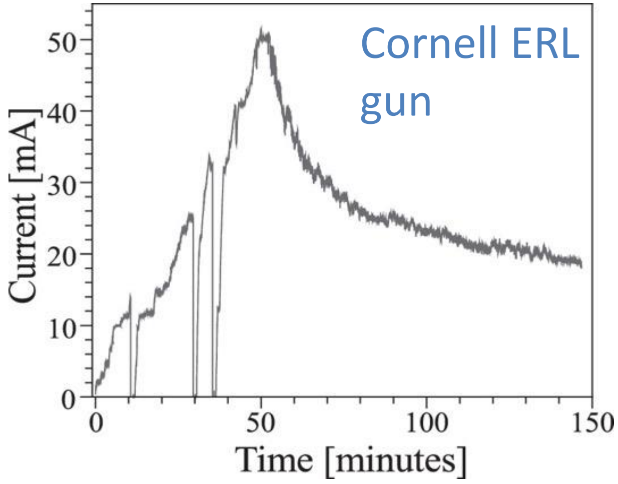


Several mA

PEPPo



LeHC 20 mA

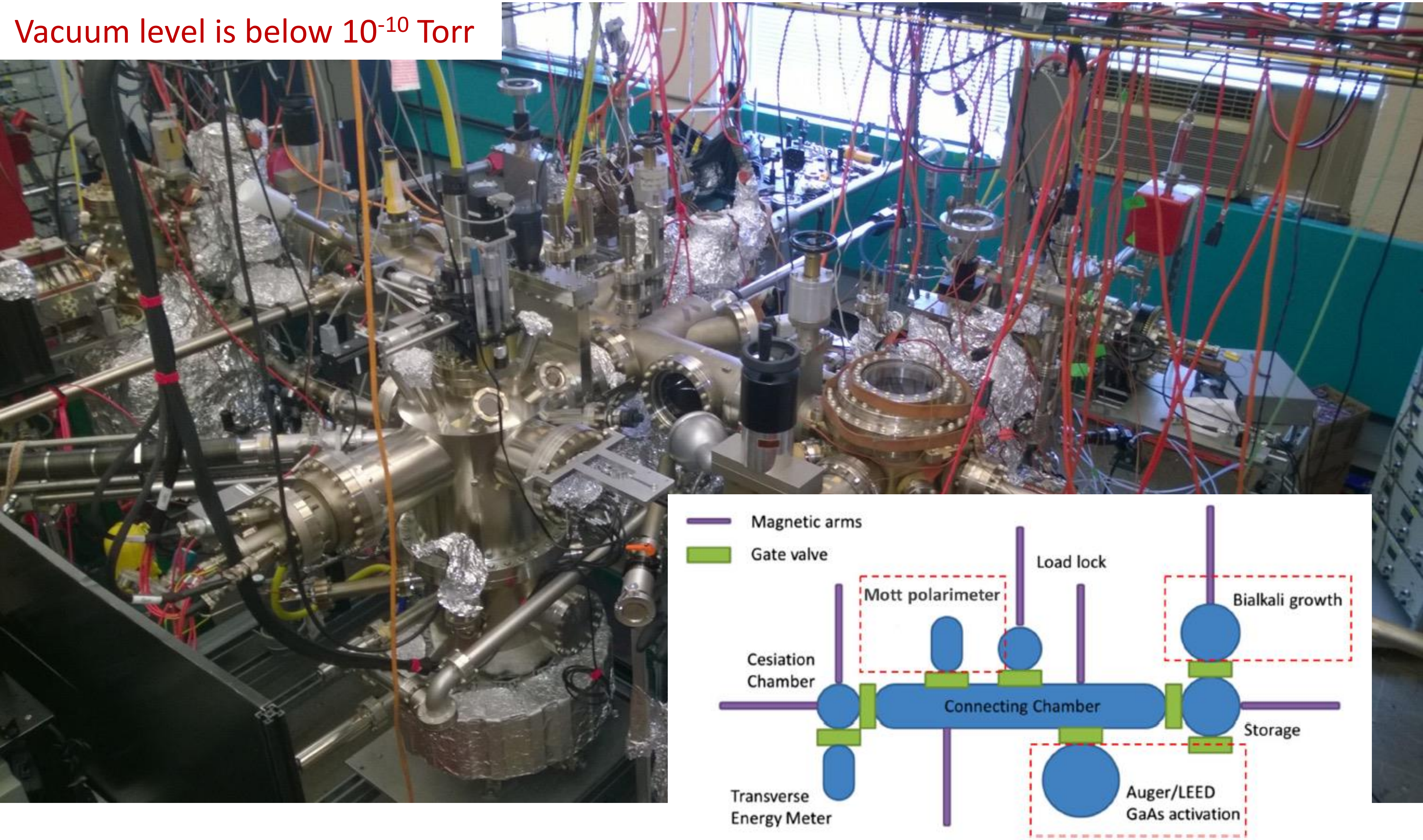


Cornell ERL gun

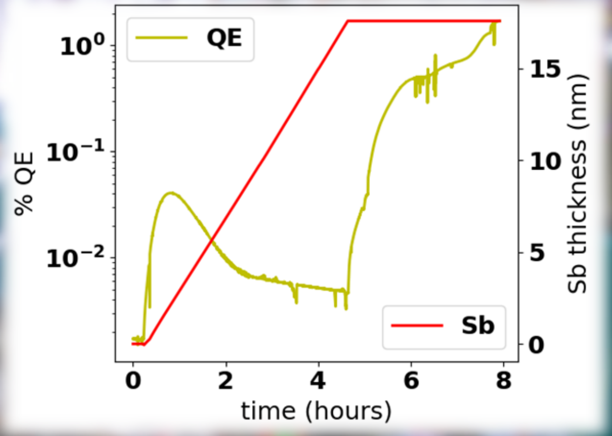
B. Dunham et al, Appl. Phys. Lett. **102**, 034105 (2013)

Cornell Photocathode Lab

Vacuum level is below 10^{-10} Torr

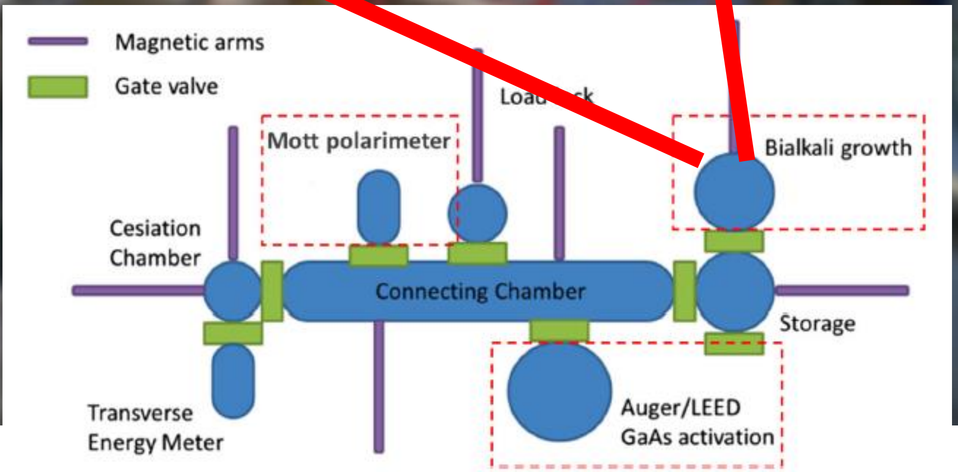


Cornell Photocathode Lab



Bialkali growth chamber

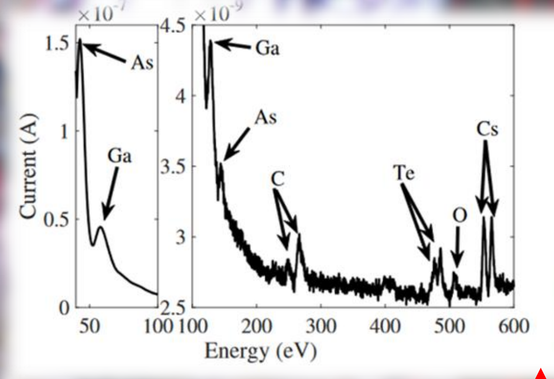
- Cs,Sb,Te sources+O_2 leak valve, thermally regulated with pneumatic valve control
- Quartz microbalance (QMB) for deposition monitoring
- Substrate heater

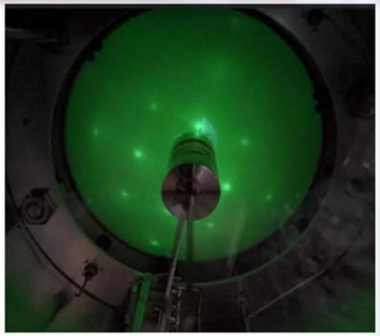


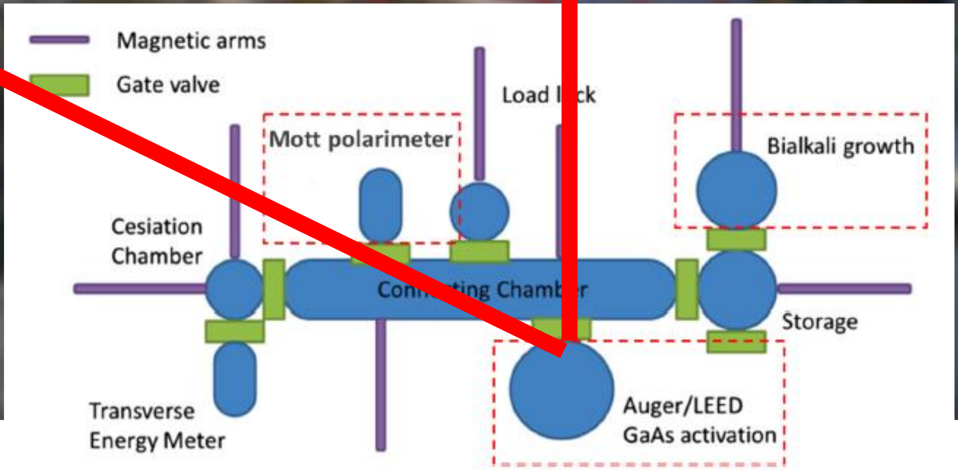
Cornell Photocathode Lab

AUGER Chamber

- Auger Electron Spectroscopy-To characterize surface chemistry
- Low Energy Electron Diffraction-Chararacterize surface crystallinity
- Cs, O$_2$ leak valve.
- Insulated, bias-able cathode holder



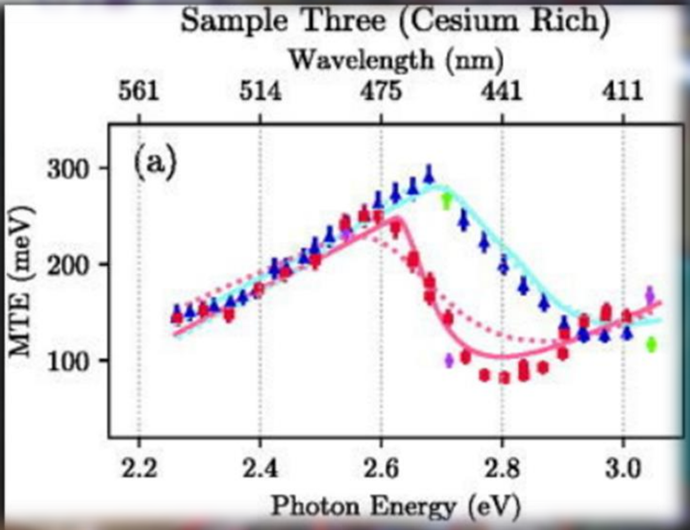


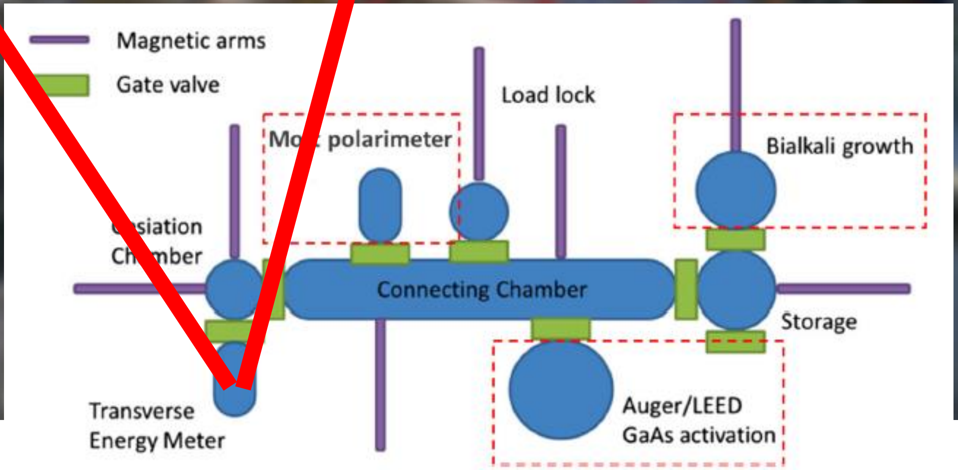


Cornell Photocathode Lab

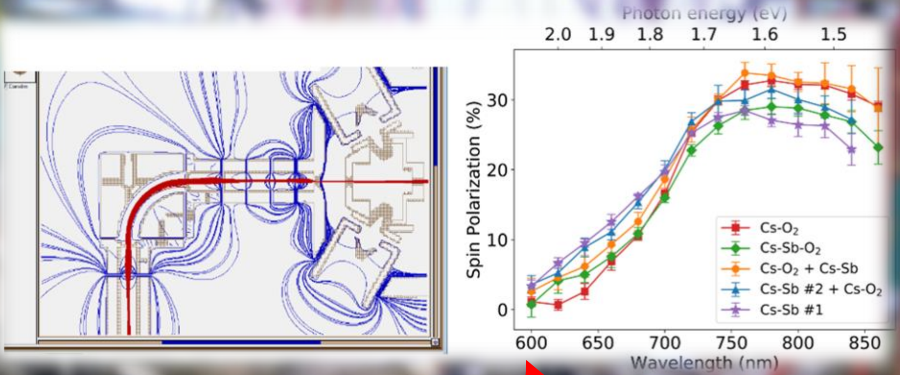
TE Meter

- 10 kV electron gun
- Microchannel plate for beam imaging
- Measure Mean Transverse Energy (MTE)



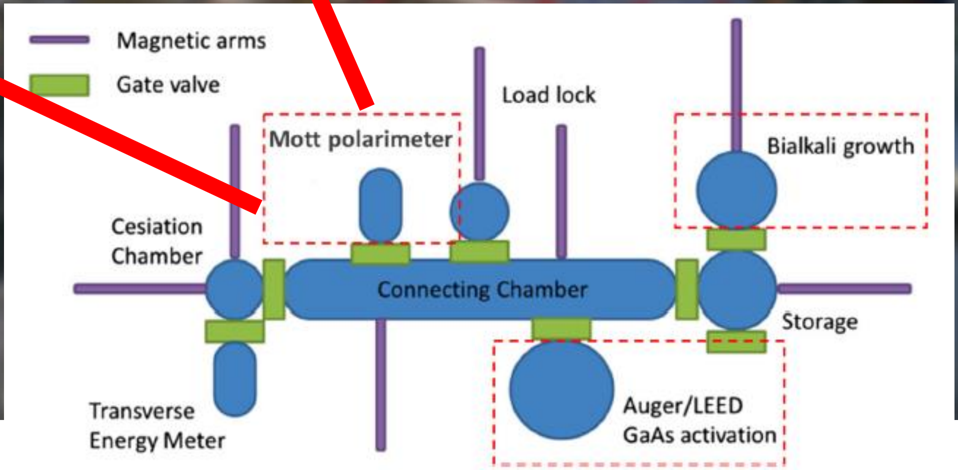


Cornell Photocathode Lab

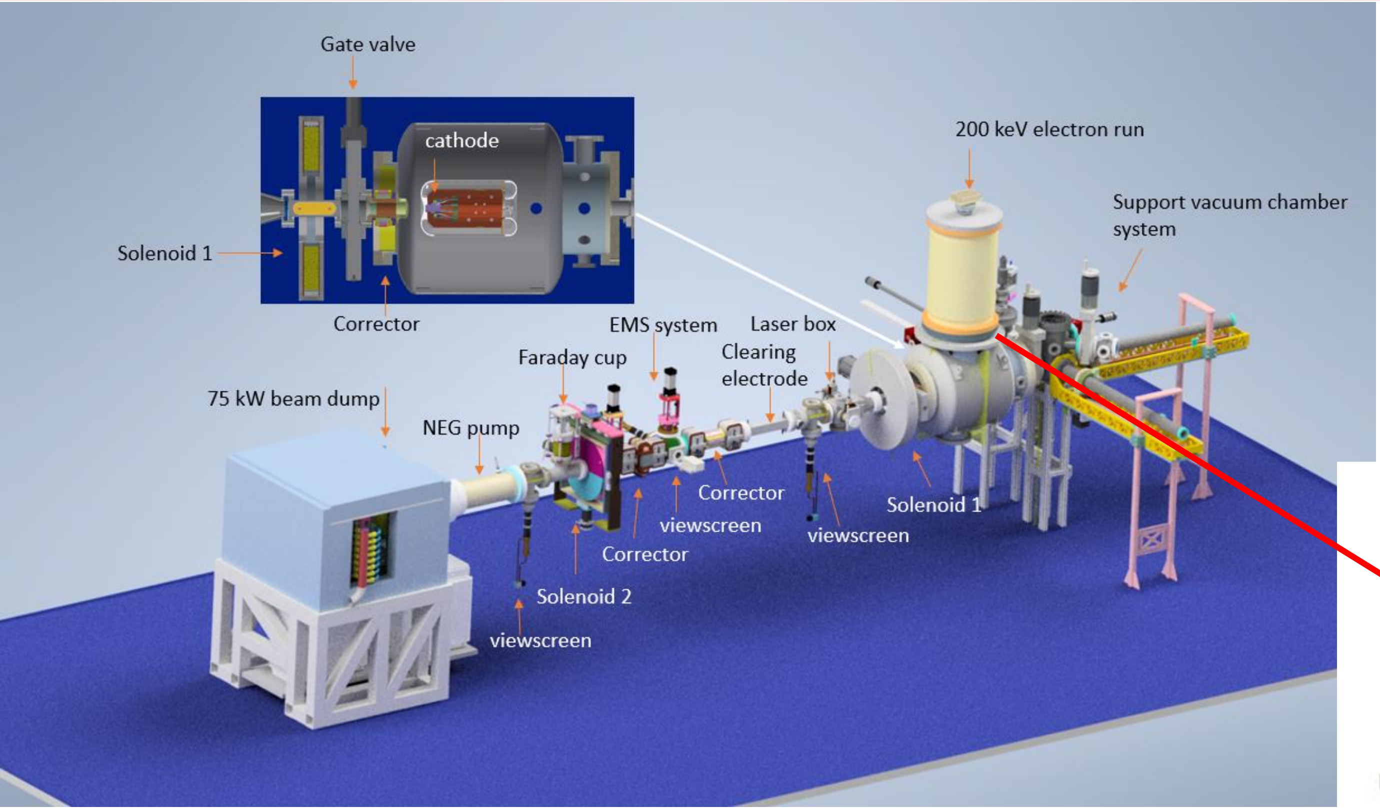


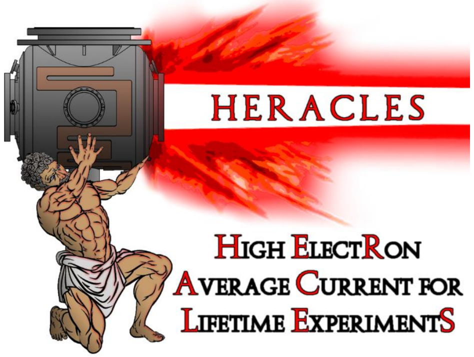
Mott polarimeter

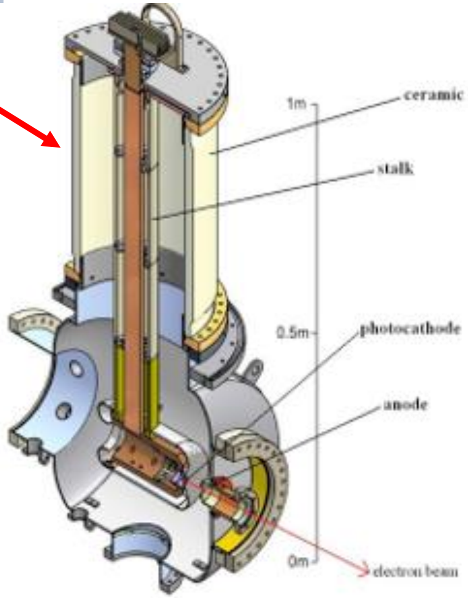
- 20 keV target bias + Einzel lens beamline
- Tungsten scattering target
- Measure spin-polarization



The HERACLES Beamline





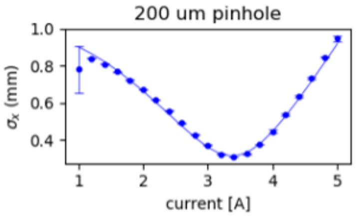


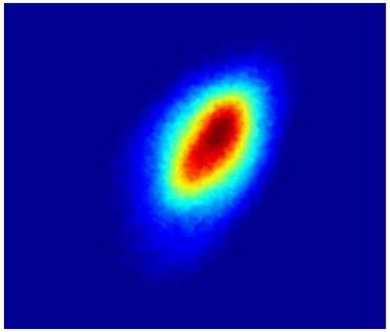
A beamline dedicated to the study of high current beam running:

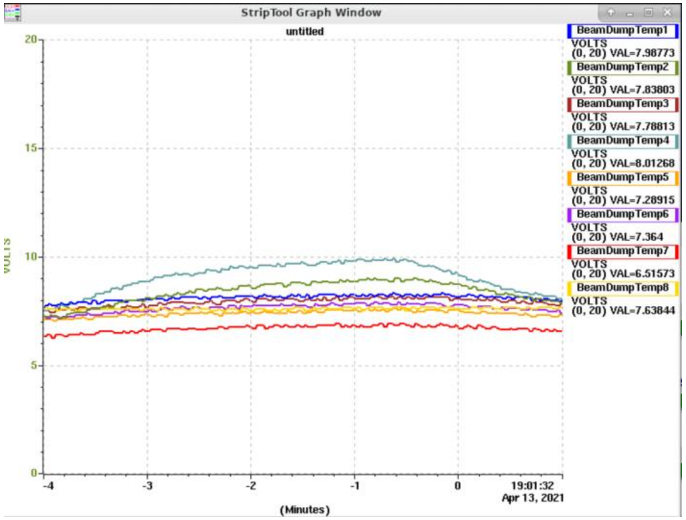
- Former CU-ERL gun 200kV @ 10 mA
- Ion clearing electrodes
- 75 kW beam dump
- EPICS based control system

Beamline and Diagnostics

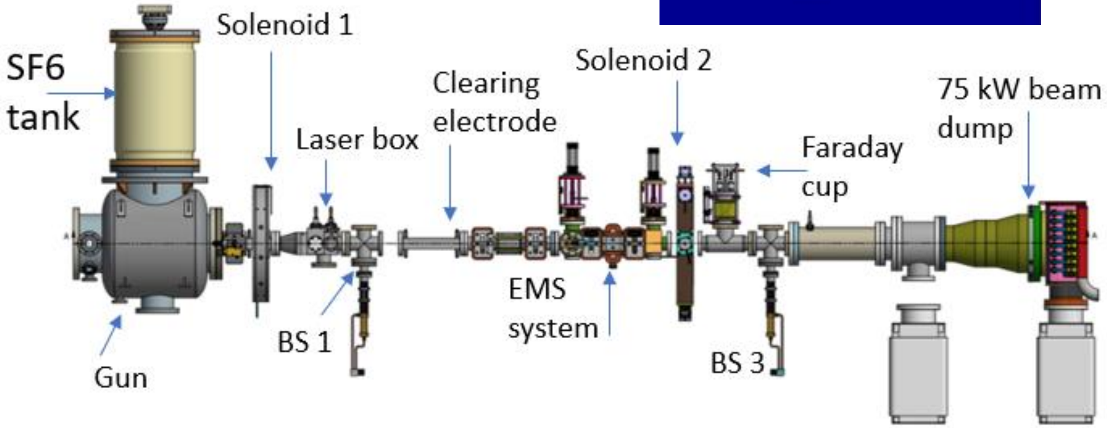
Solenoid scan characterizes cathode MTE





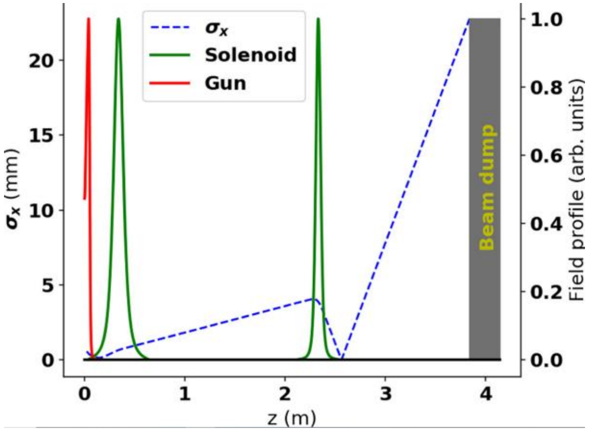


Thermal couples register beam induced temperature rise at the dump



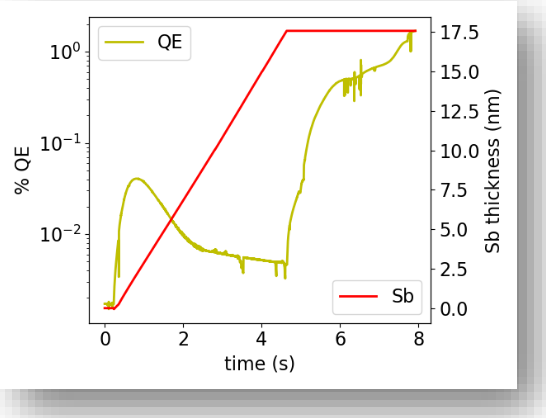
Beamline

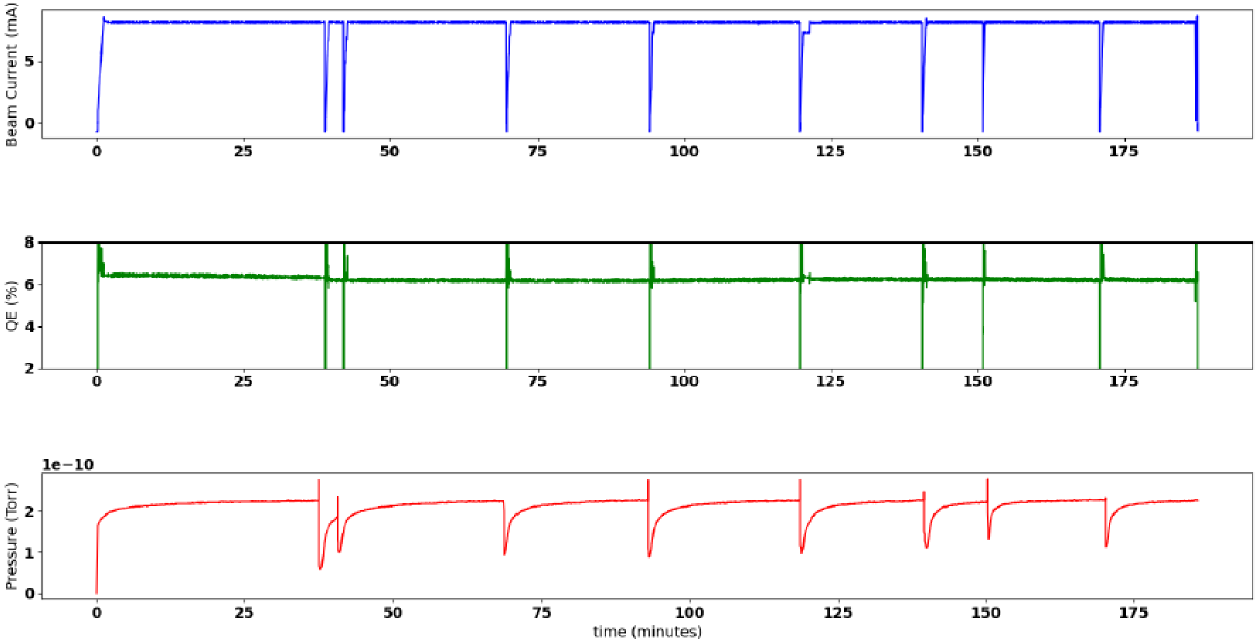
- 2 Solenoids, 3 corrector pairs (hor/vert)
- 2 clearing electrodes
- 3 screens, 1 quad detector
- 1 Faraday cup
- EMS system (not implemented)



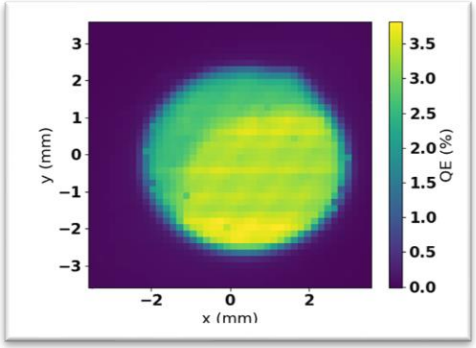
GPT simulation of fields/beam envelope

HERACLES high current performance





- Cs_3Sb cathode on stainless steel puck
- 10 mA max current, limited by radiation trips
- 8 mA constant current for 3 + hours with no significant change in cathode QE

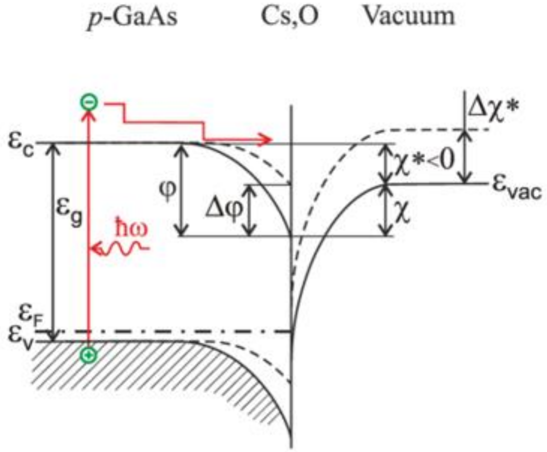


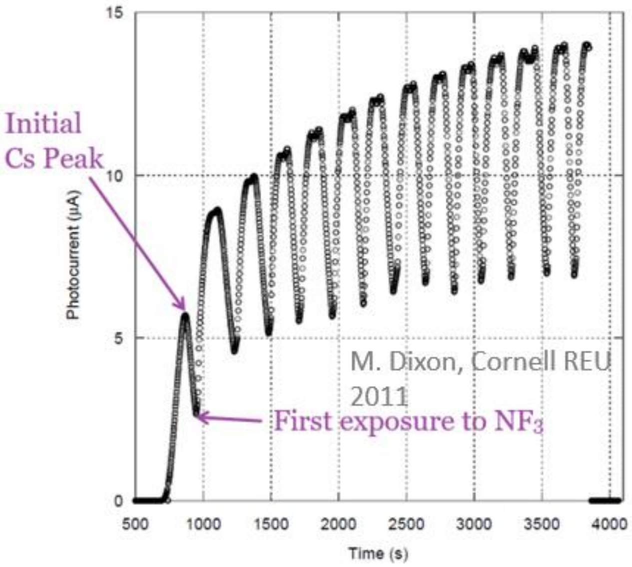
M. Andorf et al. NIMA: **1052** 168240 (**2023**) https://doi.org/10.1016/j.nima.2023.168240

Negative Electron Affinity activation of GaAs

- For high QE, GaAs is “activated” to Negative Electron Affinity (NEA)
  - NEA means the bulk conduction band minimum is larger than the vacuum level
- NEA is typically achieved by depositing a monolayer of Cs-Oxidant onto the GaAs surface can be done by:
  - **Co-deposition**: Cs is deposited until QE peaks, at which point an oxidant is leaked while Cs source remains on
  - **Yo-yo method**: the cathode is over cesiated at which point the cesium source turns off. It is then exposed to an oxidant until the QE peaks, the oxidant is then turned off and cesium source back on. The cycle is repeated numerous times.
- Either O_2 and NF_3 can be used for an oxidant
- Problem: A monolayer is a fragile thing!
  - Chemical poisoning: Interaction with residual gas
  - Ion back bombardment: residual gas is ionized and accelerated towards the cathode

V. Khoroshilov at al. (2020). Journal of Physics: Conference Series. **1482**. 012013
DOI: 10.1088/1742-6596/1482/1/012013.

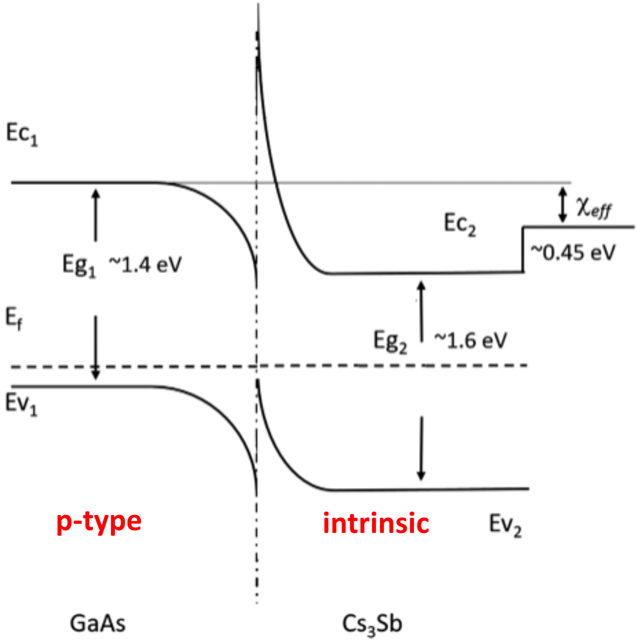


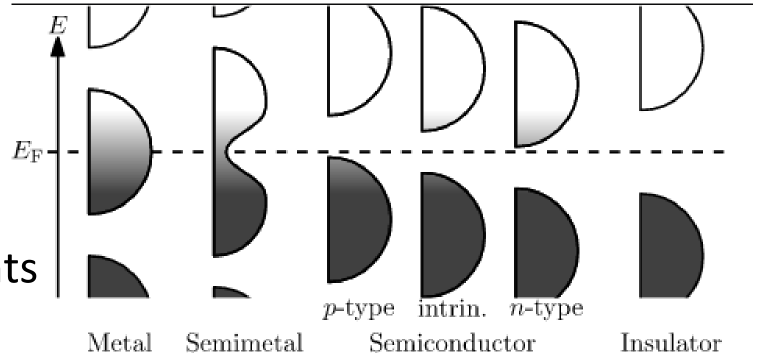


M. Dixon, Cornell REU 2011

GaAs with alternative activation layers

- NEA GaAs can be generated by forming a heterojunction at the surface with another semiconductor (activation layer) subject to two criteria:
  1. In the activation layer, the gap from the fermi level to vacuum should be **smaller** than GaAs's bandgap (~1.4 eV).
  2. So that photoemission does not occur from activation layer, its bandgap should be **larger** than GaAs's.
     - This criteria can be violated if the activation layer is sufficiently thin
- The above suggest using p-doped GaAs with an intrinsic semiconductor with a small electron affinity for the activation layer
- Cs_3Sb and Cs_2Te can form the NEA heterojunction
  - Known to be less sensitive to chemical poisoning
  - As a photoemitters they are robust at high currents
  - **This indicates Cs-Sb Cs-Te may be a very robust activation layers!**

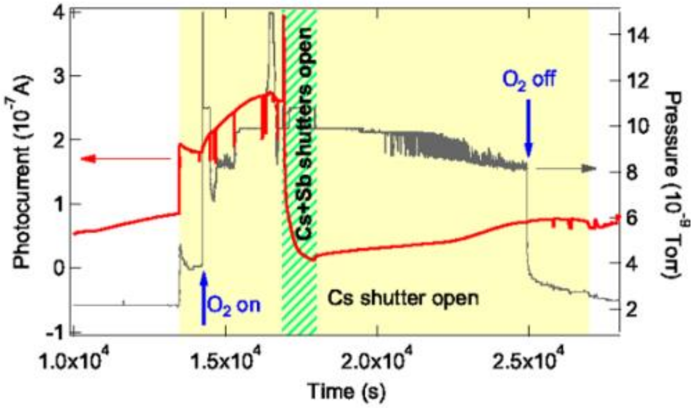


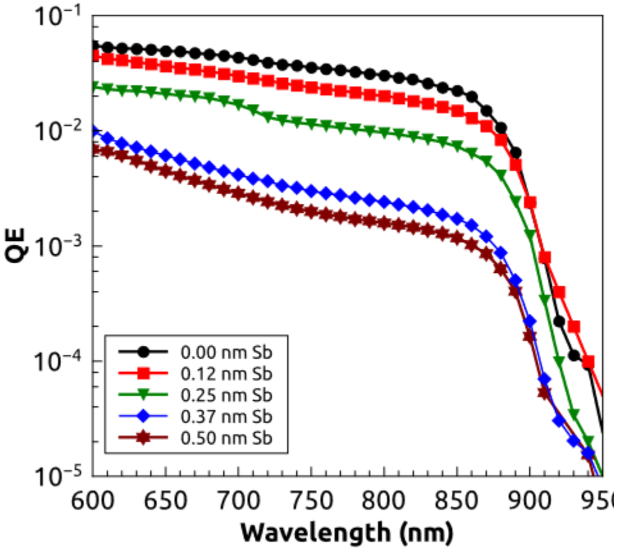


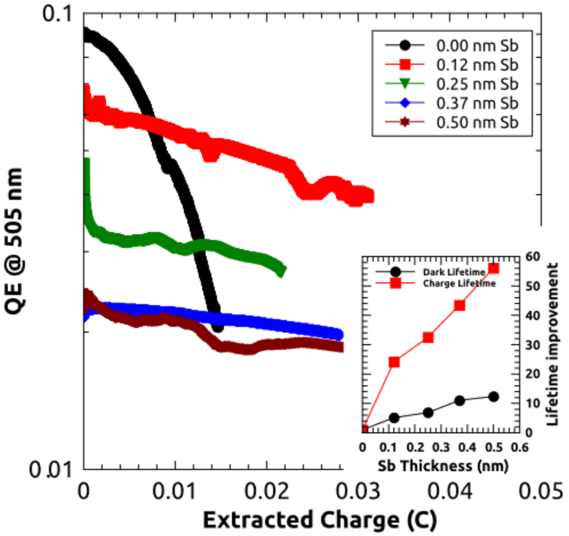
GaAs with Cs-Sb-O NEA activation layer

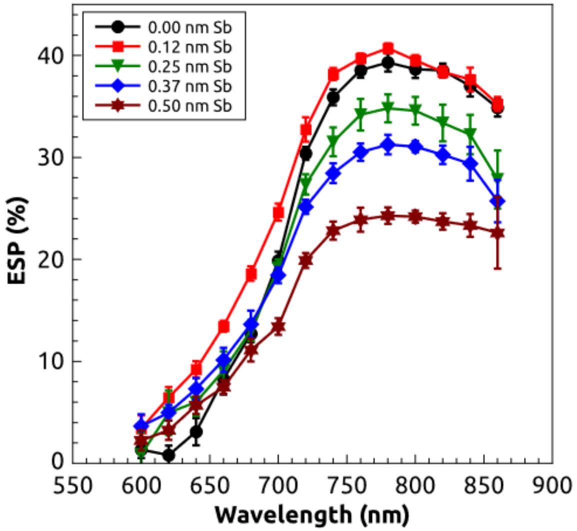
Cs-O-Sb co-deposition recipe optimized through scanning the Sb layer thickness:

- QE and electron spin-polarization decrease with Sb layer thickness
- **0.12 nm layer results in only a small QE decrease while preserving max spin-polarization!**







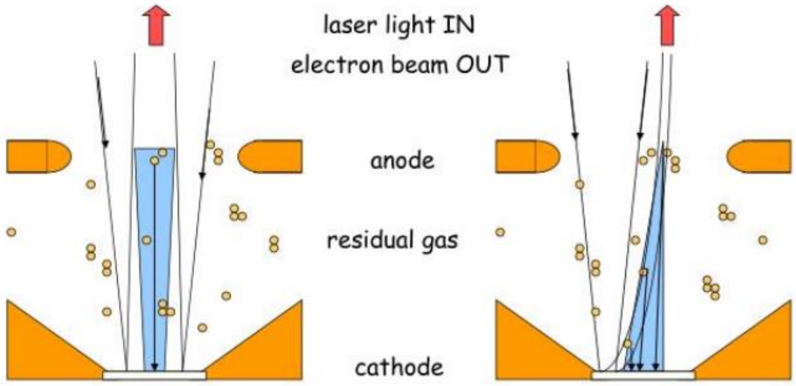


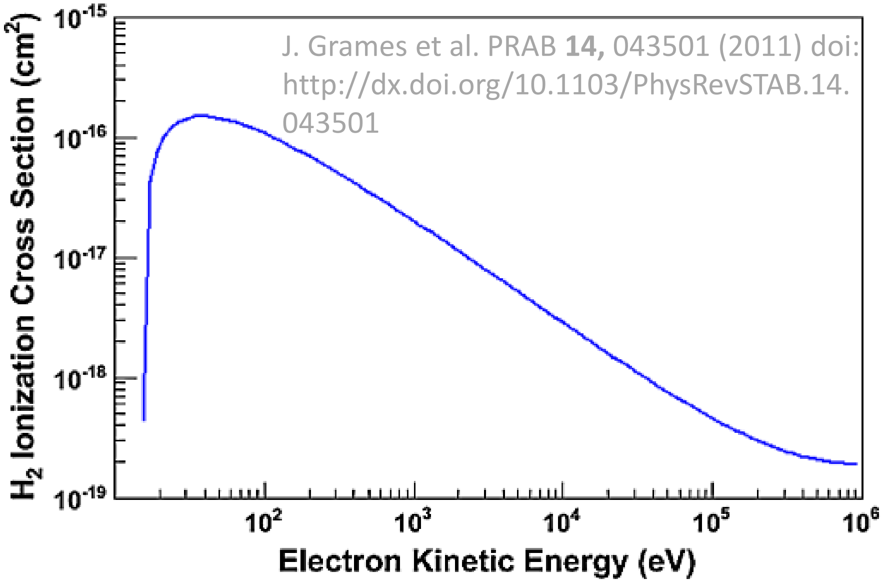
Cultrera et al, "Long lifetime polarized electron beam production from negative electron affinity GaAs activated with Sb-Cs-O: Trade-offs between efficiency, spin polarization, and lifetime (2020)"

Ion back bombardment

- An electron beam can ionize residual gas which will be positively charged
- Ions will be accelerated towards the cathode and cause damage
- So far, results have been in growth chamber
  - Low current (1-100 nA)
  - Low voltage (-18 V)
  - Main source of degradation comes from vacuum poisoning
- In a high voltage DC gun, induced damage from ion back bombardment is more severe
  - Lower energy ions can sputter off activation layer
  - High energy ions damage GaAs crystal structure
- **Operation at high voltage and current is critical to testing efficacy of alternative activation layers!**

Image courtesy J. Grames





J. Grames et al. PRAB **14,** 043501 (2011) doi: http://dx.doi.org/10.1103/PhysRevSTAB.14.043501

Cs-Sb-O activated GaAs cathode in HERACLES

Goal

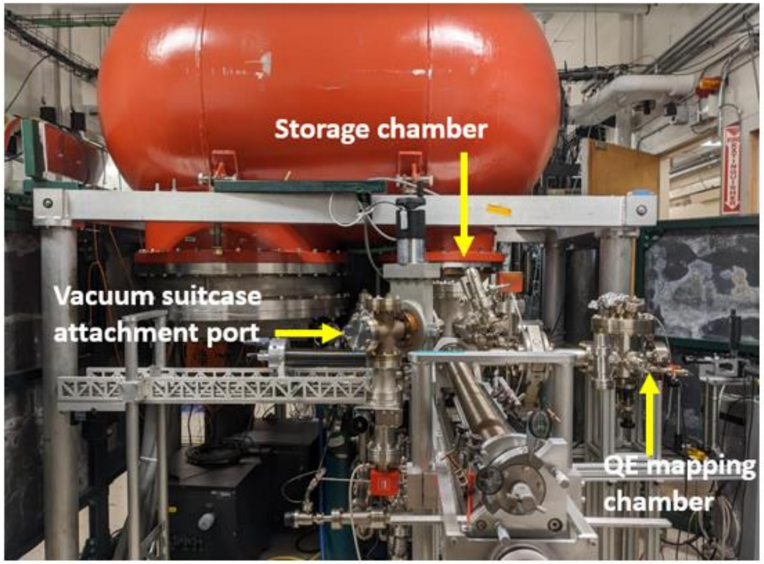
- Compare the charge lifetimes of Cs-Sb-O vs Cs-O activated GaAs.

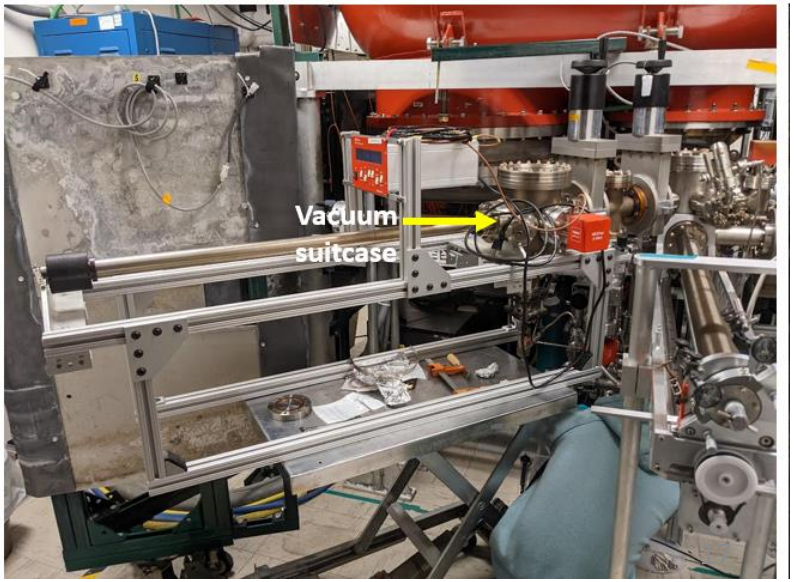
Sample Preparation:

- Cs-O:GaAs
    - activated with co-deposition in a chamber attached to the rear of HERACLES.
    - Lifetime measurements performed within hours of activation
- Cs-O-Sb:GaAs
    - Activated in Mother chamber and transported via a vacuum suitcase to HERACLES
    - ~24 hours between activation and lifetime measurement

Run Conditions

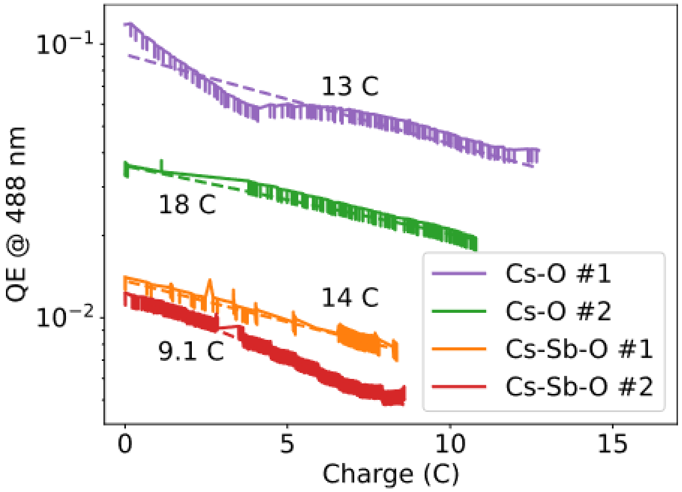
- 1 mA beam current held constant for 2-3 hours
- 488 nm drive laser. QE map at 532 and 780 nm
- Typical vacuum pressure in the low e-10's during run

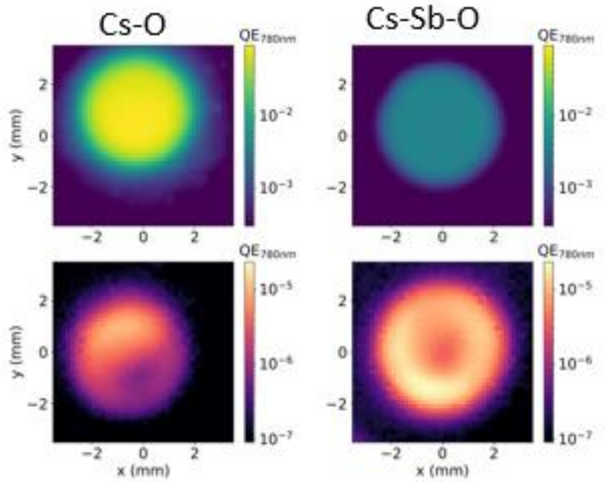


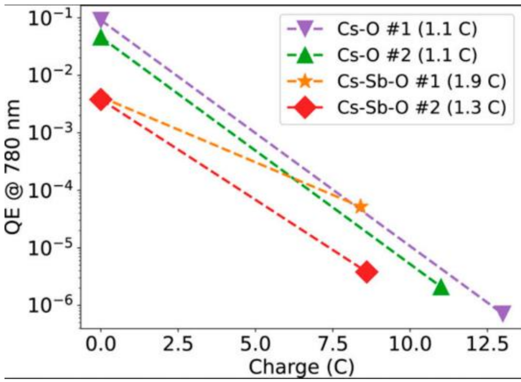


Cs-Sb-O activated GaAs cathode in HERACLES (results)

- We did not see an improvement in the charge lifetime at 488 nm
- Pre/post run comparisons of QE indicate lifetime at 780 nm may have improved.
  - Relative QE change smaller for Cs-Sb-O samples
- Cs-Sb-O had lower QE’s
  - We observed roughly a factor of 3 reduction in the QE between activation in the Mother chamber and beginning the run.
  - Required laser power a factor of 4-10 higher
- **The story is not over!**
  - **New growth chamber on HERACLES to support Cs-Sb-O/Cs-Te-O activation**
  - **780 nm drive laser**



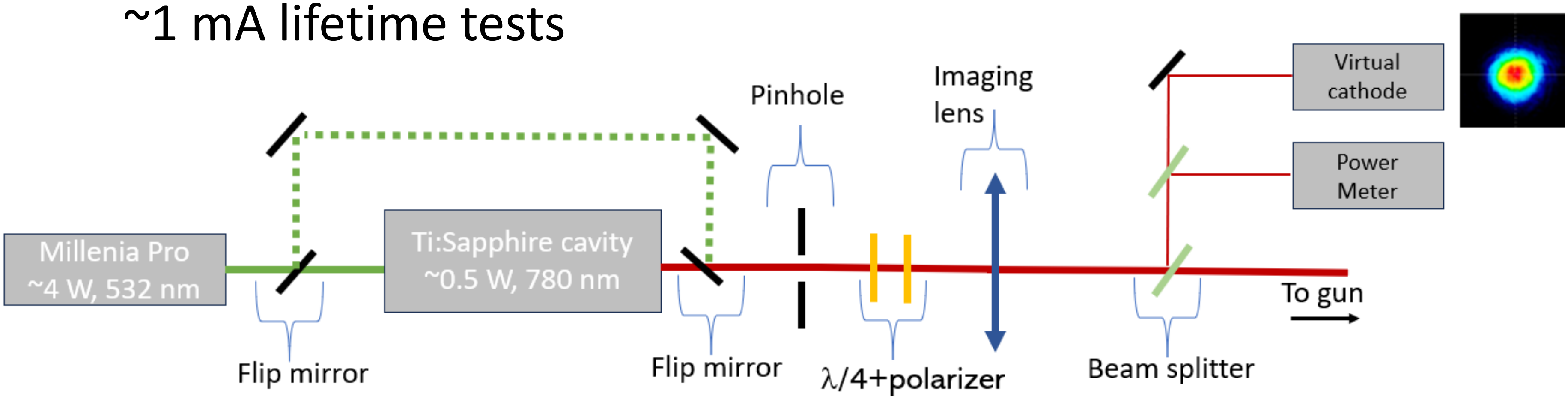




HERACLES DRIVE LASER

New Drive Laser for high current GaAs operation!

- Removed argon laser ~488 or 360 nm
- Ti:Sapphire oscillator: lasing at GaAs bandgap energy
- 0.5 W output, suitable for ~1 mA lifetime tests



New Growth Chamber!

- Replaced previous chamber (Cs, +QE mapping)
- New chamber:
    - Sources: Cs, Sb, Te, O_2
    - Instrumentation: QMB, sample heater, QE map, Lock-in.
    - Labview VI: QE monitoring, Source ramping, Source (flux) calibration

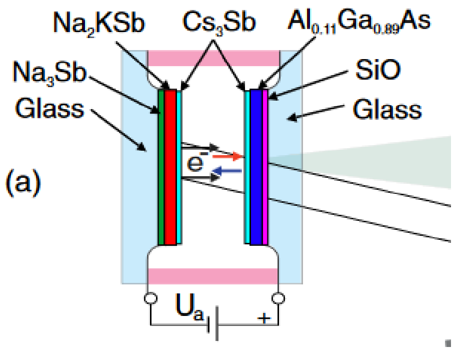
Status:

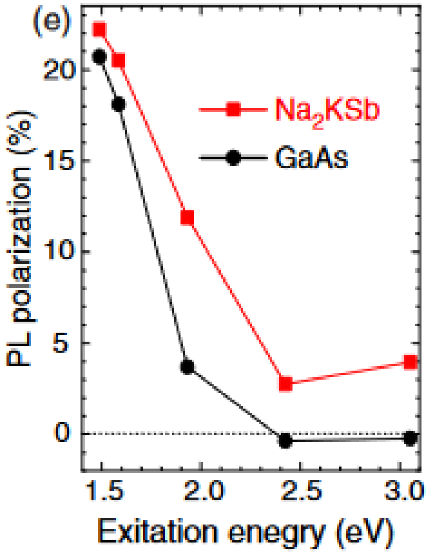
- Installed on HERACLES
- UHV achieved with sources cold
- Sources calibrated
- Growth

ESP from an Alkali-antimonide?

Rusetsky et al. reported spin polarized photoemission alkali antimonide

- Cs_3Sb NEA layer on Na_2KSb
- Photoluminescence (PL) $Al_{0.11}Ga_{0.89}As$ detector
- Circular PL degree correlated to ESP
- Calibrated with an NEA GaAs photocathode





Alkali-antimonides are robust, efficient emitters. Great opportunity for developing a robust spin polarized electron source!

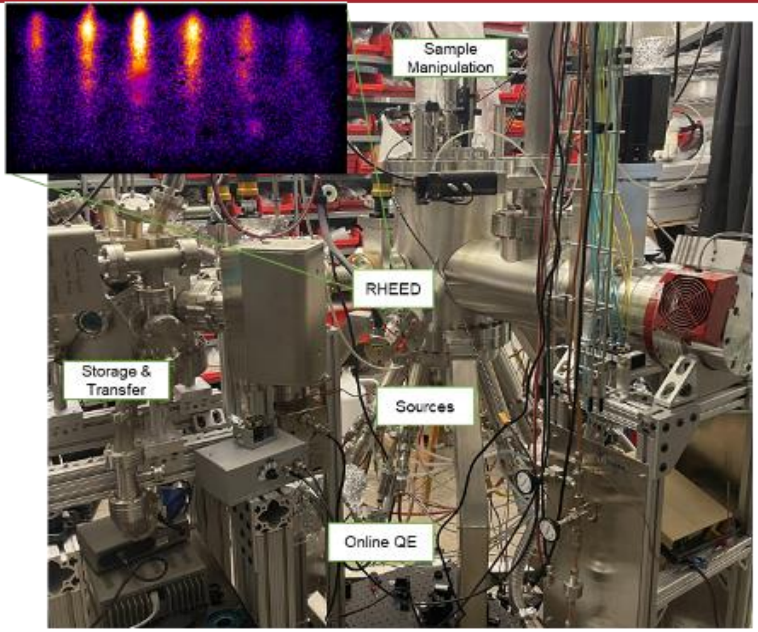
PHYSICAL REVIEW LETTERS **129,** 166802 (2022)

Editors' Suggestion

New Spin-Polarized Electron Source Based on Alkali Antimonide Photocathode

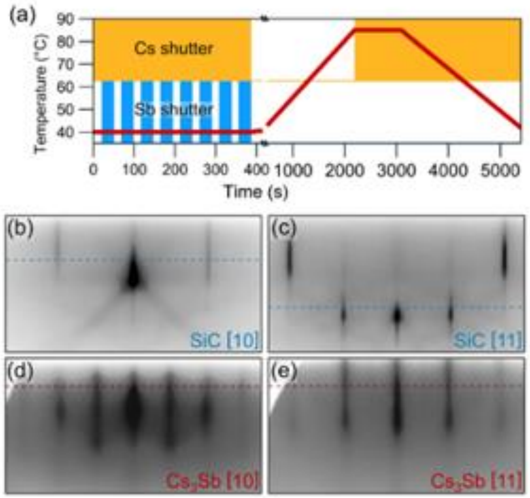
V. S. Rusetsky,[1,2] V. A. Golyashov,[1,3,4] S. V. Eremeev,[5] D. A. Kustov,[1] I. P. Rusinov,[6] T. S. Shamirzaev,[1,4]
A. V. Mironov,[2] A. Yu. Demin,[2] and O. E. Tereshchenko[1,3,4,*]

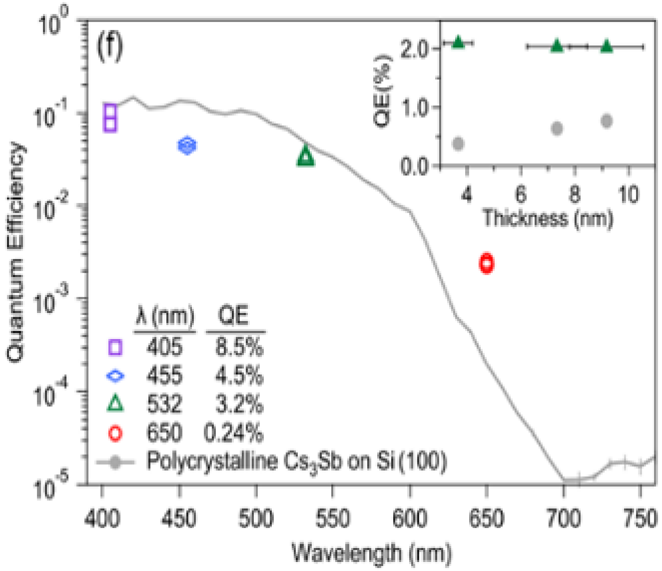
Molecular Beam Epitaxy in BBL



MBE system in BBL:

- Growth parameters monitored with RHEED—standard diagnostic for *epitaxy*
- Single crystal epitaxial growth of Cs_3Sb!
  - Thin, ~10 nm, but still efficient photoemitter

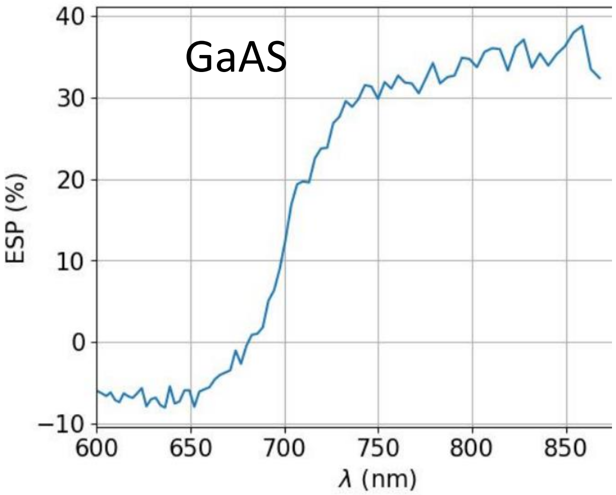


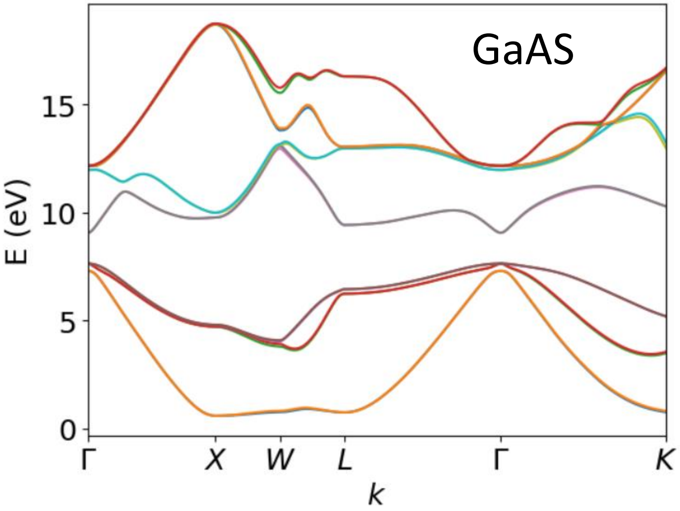


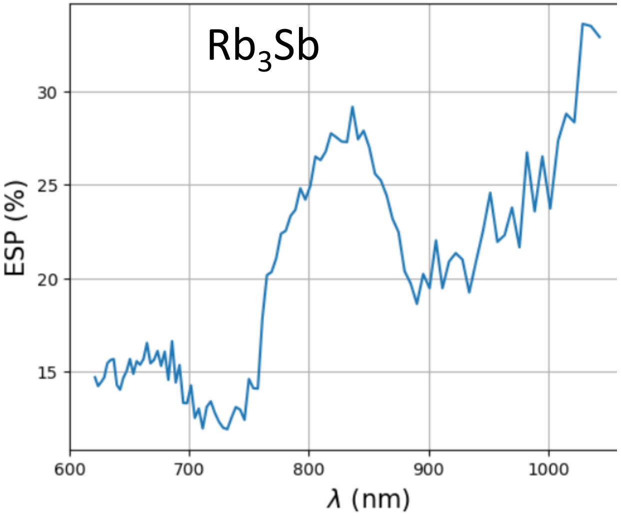
DFT of Rb_3Sb

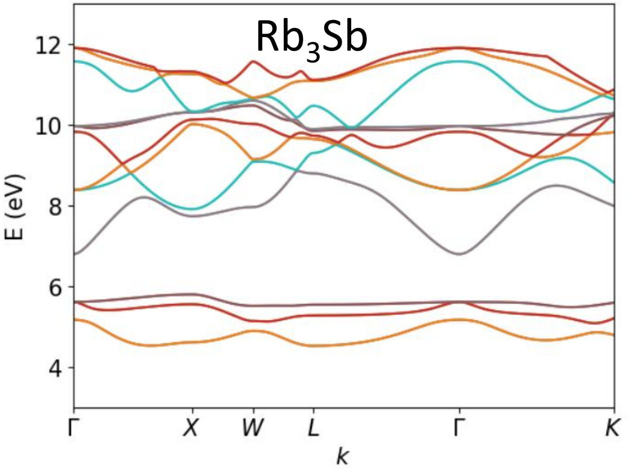
With Tomás Arias, Tyler Wu, Zahin Ritee (REU), Cornell

- *Ab initio* DFT simulations using jDFTx for predicting ESP
- We assume NEA and do not account depolarization effects
- Reasonable predictions with GaAs
- The case for Rb_3Sb looks optimistic!
  - Band degeneracy at Γ point lifted by SOC
  - Predicts ESP









Budget and Deliverables

	FY2023	FY2024	Total
a) Funds allocated	230 k	180 k	410
b) Actual cost to date	120 k	5 k	125 k

2023/2024 timeline

Calendar year 2023/2024	1st quarter	2nd quarter	3rd quarter	4th quarter
Growth chamber testing	🟧	🟩	🟩	🟩
GaAs Cs-Sb Lifetime	🟧	🟧	🟧	🟩
GaAs Cs-Te lifetime			🟧	🟧
Rb_3Sb DFT study	🟧	🟩	🟩	🟩
Rb_3Sb growth/characterization			🟧	🟧

Task Active 🟧

Projected complete 🟩

Thank you for your attention!

BACKUP SLIDES

Tasks and milestones

Task 1: *Design, fabricate, install, and test a new growth chamber for CsSb and CsTe NEA activation of GaAs and storage chamber for HERACLES.*

The new chambers will replace the existing storage chamber and QE mapping chamber on HERACLES. The new storage chamber will increase the storage capacity from 3 to 5 cathodes. Because the new growth chamber is directly attached to HERACLES it will eliminate the need for a vacuum suitcase transfer of activated samples, dramatically reducing the cathode preparation time and thereby increasing the throughput of our cathode testing abilities. In addition to reduced preparation time, there is reduced time between growth and high current measurements, resulting in a more accurate measurement of the charge lifetime.

Milestone 1.1: The new chambers are installed on HERACLES, attached to the gun gate valve and existing venting and oxygen lines, UHV is achieved and the growth chamber's instrumentation (source heaters, sample heater, lock-in amplifier, quartz microbalance) are tested.

Milestone 1.2: A Growth of Cs-Sb and Cs-Te are performed. QE and charge lifetime measurements at low current and voltage are performed in the chamber to benchmark the new growth chamber against prior results obtained in our photocathode laboratory.

Tasks and milestones

Task 2: *Perform the systematic study of Cs_3Sb NEA activated GaAs lifetime at mA scale beam current in HERACLES: vary Cs_3Sb thickness and illumination laser wavelength to (i) determine the optimized charge lifetime and (ii) disentangle the degradation mechanisms.*

There is a trade-off between lifetime and quantum efficiency for Cs_3Sb NEA activated GaAs. The measurements from that study were performed in a growth chamber at low current and voltage. The environment of a high voltage (200 keV) electron gun at 1 mA beam current is much harsher with significantly more ion-back bombardment and laser induced thermal degradation. By measuring the lifetime as a function of Cs-Sb thickness *in situ* the efficacy of the activation layer, and its optimized value can be determined. The cathodes quantum efficiency can be a factor of 2-3 higher when operated with visible light photons vs infrared. By measuring the charge lifetime at the same beam current and gun voltage, but different photon energies (and therefore, different laser powers) the effect of laser induced thermal degradation can be determined. Disentangling the *in-situ* degradation methods is a first step towards engineering long lifetime, spin-polarized photocathodes.

Milestone 2.1 Procure the visible and NIR lasers and modify our injector laser optics to enable quick switching between visible and IR illumination. At low current (~100 nA) the beam profile will be observed, and the linear optics (solenoid strengths, correctors) adjusted so that the beam orbit at both wavelengths is a close as possible.

Milestone 2.2 Do the lifetime study with Cs_3Sb. We plan to scan the activation layer between 0.05 to 0.4 nm. We will repeat runs at each thickness and wavelength to ensure reliability of our results.

Tasks and milestones

Task 3: *Perform the systematic study of Cs_2Te NEA activated GaAs lifetime at mA scale beam current in HERACLES: vary Cs_3Sb thickness and illumination laser wavelength to (i) determine the optimized charge lifetime and (ii) disentangle the degradation mechanisms.*

Similar to Task 2, we will vary the thickness activation layer thickness and laser illumination wavelength while measuring the charge lifetime in HERACLES. More generally, the Cs_2Te activation layer can be made thicker while still yielding percent level QE. Again, by performing the charge lifetime measurements *in situ* and with different illumination wavelengths, the optimized activation layer can be found, and the degradation mechanisms assessed. Furthermore, by comparing this task's results to task 2 we not only determine which activation layer performs better but gain insight into how the activation layer properties (for example, binding energy to the GaAs surface) affects charge lifetime. This information is another step toward engineering long-lifetime, robust spin-polarized photocathodes.

Milestone 3.1 Do the lifetime study with Cs-Te. We plan to scan the activation layer between 0.5 to 1.2 nm. We will repeat runs at each thickness and wavelength to ensure reliability of our results.

Tasks and Milestones

Task 4: *Perform a comprehensive computational study of cubic-phase monoalkali antimonides to determine its ultimate theoretical performance as spin-polarized electron source*

The possibility of using an epitaxially grown alkali-antimonide as a novel spin-polarization source is a fascinating prospect. Multi-crystalline alkali-antimonide photocathodes have been well documented as being able to reliably deliver 10s of mA beam current for several hours without performance degradation. Can this robustness be translated to a robust spin-polarized cathode in the single-crystal cubic case?

Milestone 4.1: Using Density Functional Theory (DFT) we will consider several critical factors: is Rb_3Sb the optimal among monoalkali antimonides for spin polarized production? What is total depth of electron affinity modulation of a Cs-containing activation layer, such as an epitaxial layer of Cs_3Sb? To what extent is positive electron affinity permissible for spin polarized emission, and what is the ultimate theoretical spin polarization possible?

Tasks and Milestones

Task 5: *Perform epitaxially growths of alkali-antimonides to better understand their potential as a spin-polarized source*

Our first target material for epitaxial growth will be Rb_3Sb. Using our photocathode laboratory's MBE chamber, we will determine the conditions for epitaxial growth while monitoring the atomic structure using RHEED. Once the atomic structure has been successfully produced, the possibility of NEA using Cs_3Sb will be explored. The sample can be transported from the MBE to our Mott polarimeter were the degree of spin-polarization can be measured.

Milestone 5.1: In our MBE chamber, produce epitaxially grown single-crystal cubic Rb_3Sb as indicated with RHEED.

Milestone 5.2: Perform a spectral analysis of the epitaxially grown Rb_3Sb with a Cs_3Sb activation layer to determine feasibility of NEA activation.

Milestone 5.3: Measure the obtained spin-polarization of the surface activated Rb_3Sb in either our laboratory's Mott polarimeter or at the PARADIM user facility at Cornell using VLEED polarimetry.