Epitaxial growth of the cesium antimonide photocathode

Chad Pennington

Cornell University , Department of Physics + CLASSE

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The Center for BRIGHT BEAMS
A National Science Foundation Science & Technology Center

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Cornell University: Elena Echeverria, Matt Gordon, Michael Kaemingk, Tomas Arias, Jared Maxson.

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Brookhaven National Laboratory: Kali Mondal, Mengjia Gaowei.

SLAC: John Smedley.

University of Salerno, Italy: Alice Galdi.

Outline

1. Motivation for the epitaxial growth of Cs_3Sb.

2. Growth of ordered photocathodes using pulsed laser deposition (PLD) and *in-operando* x-ray characterization.

3. Measuring quantum efficiency with optical interference in the photocathode-substrate system.

4. Cs_1Sb_1: a highly oxygen resistant, visible photocathode.

5. Summary and Conclusions.

Epitaxial Growth

- Cs_3Sb photocathodes are conventionally grown polycrystalline with disordered surfaces.
- Reducing the intrinsic emittance may be limited by the surface disorder in the form of defects, grain boundaries, and roughness.

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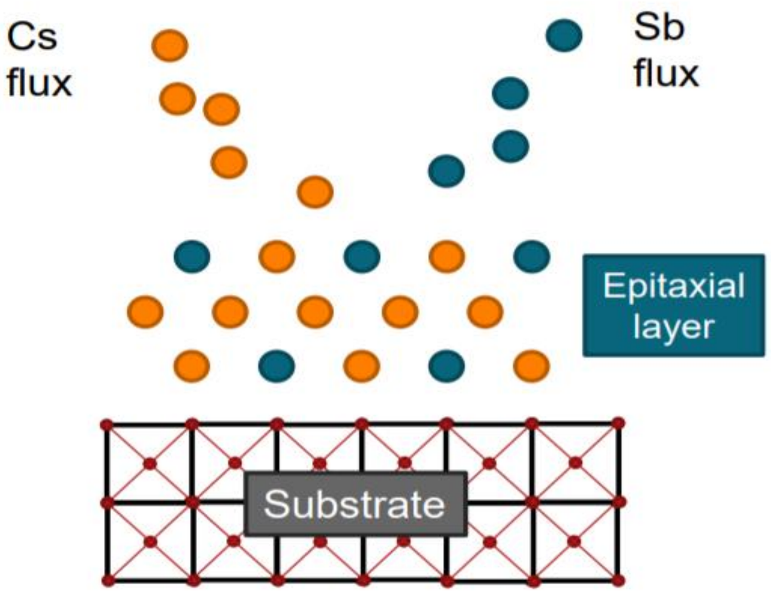
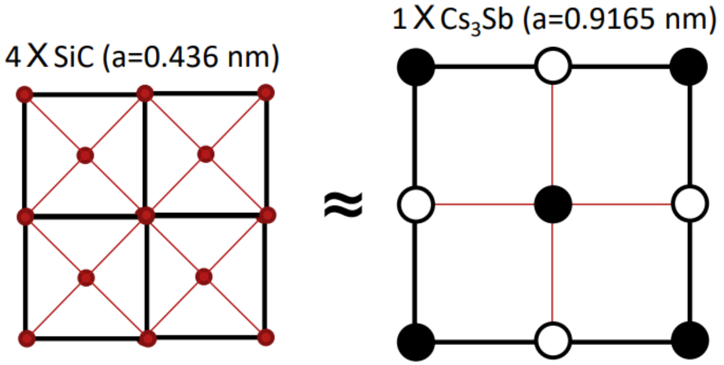
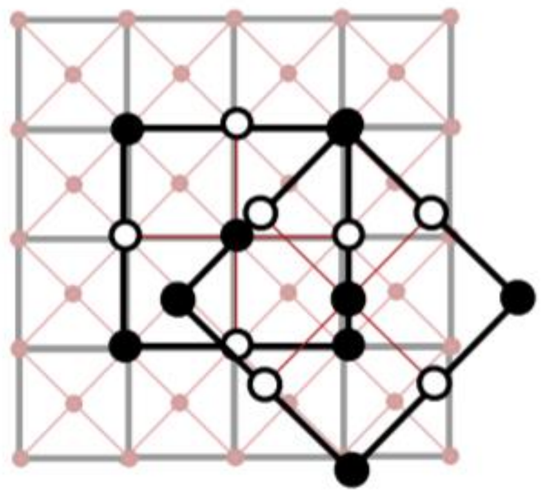


Cs_3Sb is grown on lattice matched, 3C-SiC substrates.





Why epitaxy?

- Towards higher brightness: a figure for the quality of the electron beam.

$$B_n = \frac{2m_e c^2 I}{\sigma_x^2 MTE}$$

Beam current: quantum efficiency, laser fluence

Mean transverse energy: Intrinsic momentum spread + roughness + laser heating + ...

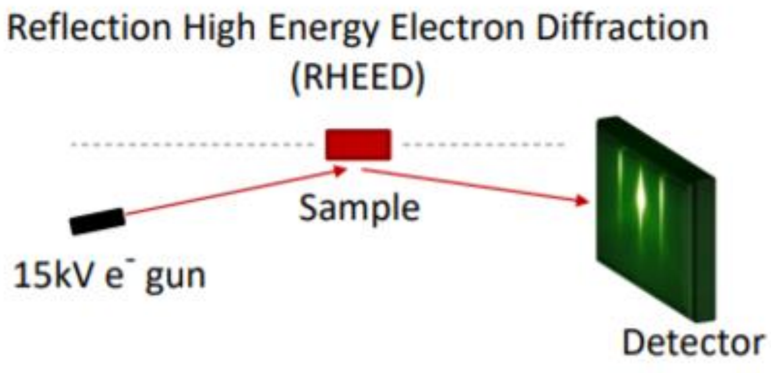
- Material ordering eliminates defects (roughness, grain boundaries) that contribute to electron momentum spread (MTE).

- Epitaxy opens the door to band structure and/or QE engineering – similar to work on single crystal GaAs and GaN.

-. W. Liu, *et.* , Appl. Phys. Lett. 109, 252104 (2016).

- J. Marini,, Polarization engineered N-polar Cs-free GaN photocathodes, J. Appl. Phys. 124, 113101 (2018).

RHEED: one tool for determining epitaxy



- RHEED identifies the surface structure through *grazing incidence* over the first few atomic layers of the sample.

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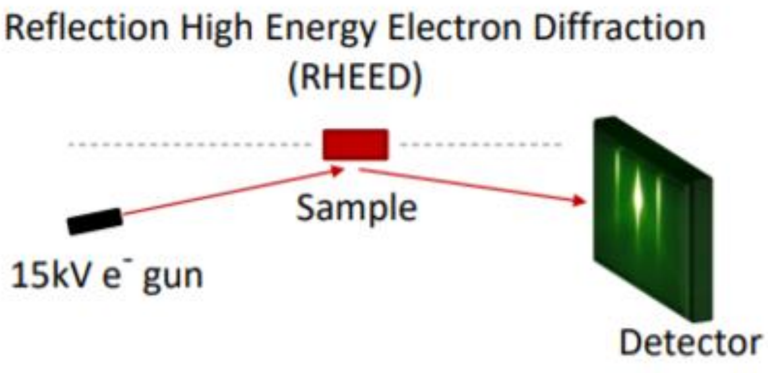

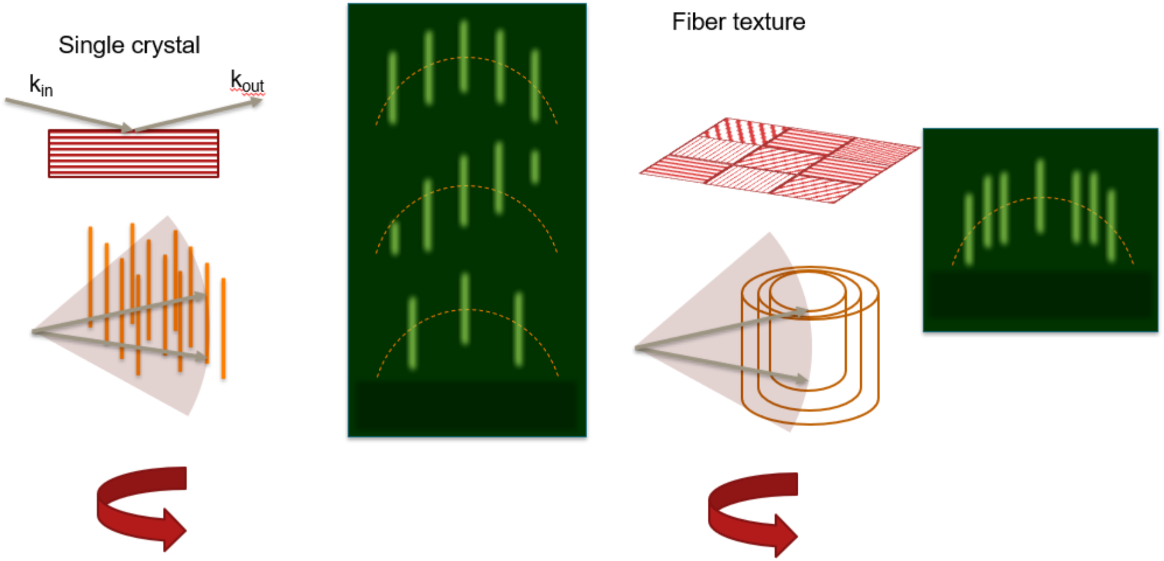

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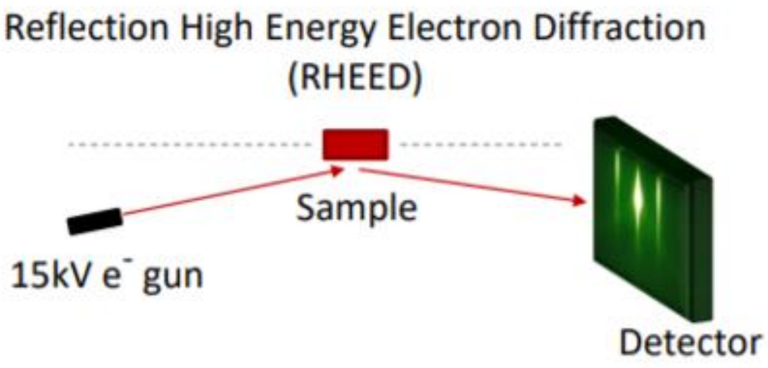

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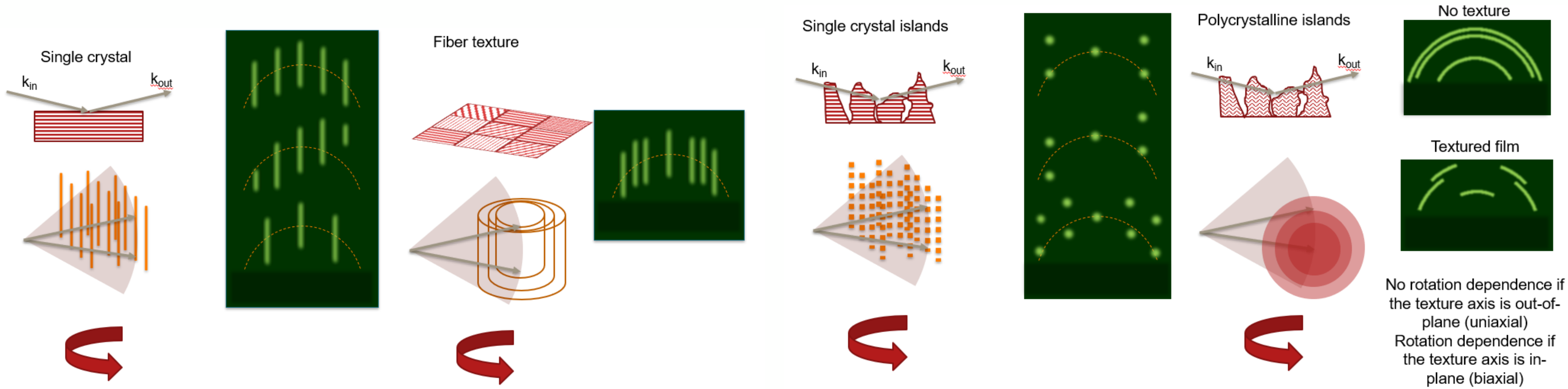
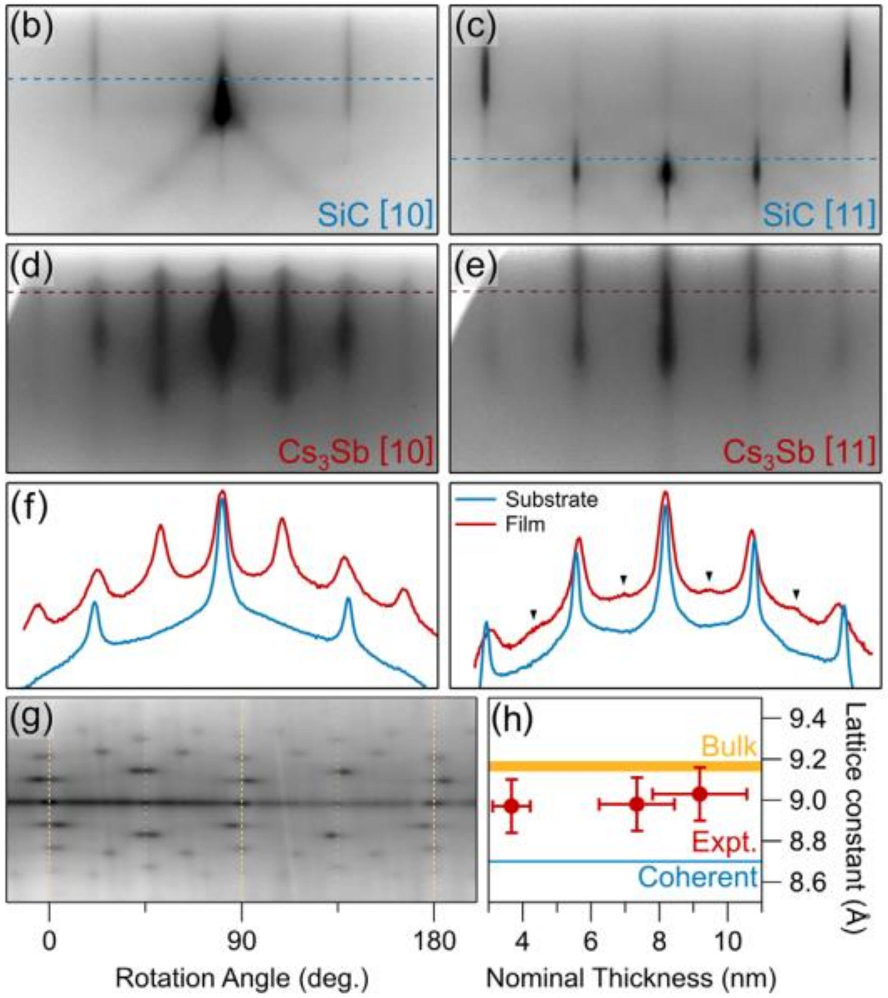


No rotation dependence if the texture axis is out-of-plane (uniaxial)
Rotation dependence if the texture axis is in-plane (biaxial)

X-ray diffraction (XRD) is another tool that can determine epitaxy.
See Kali Mondal’s talk in this session!

Results on Cs_3Sb grown with MBE

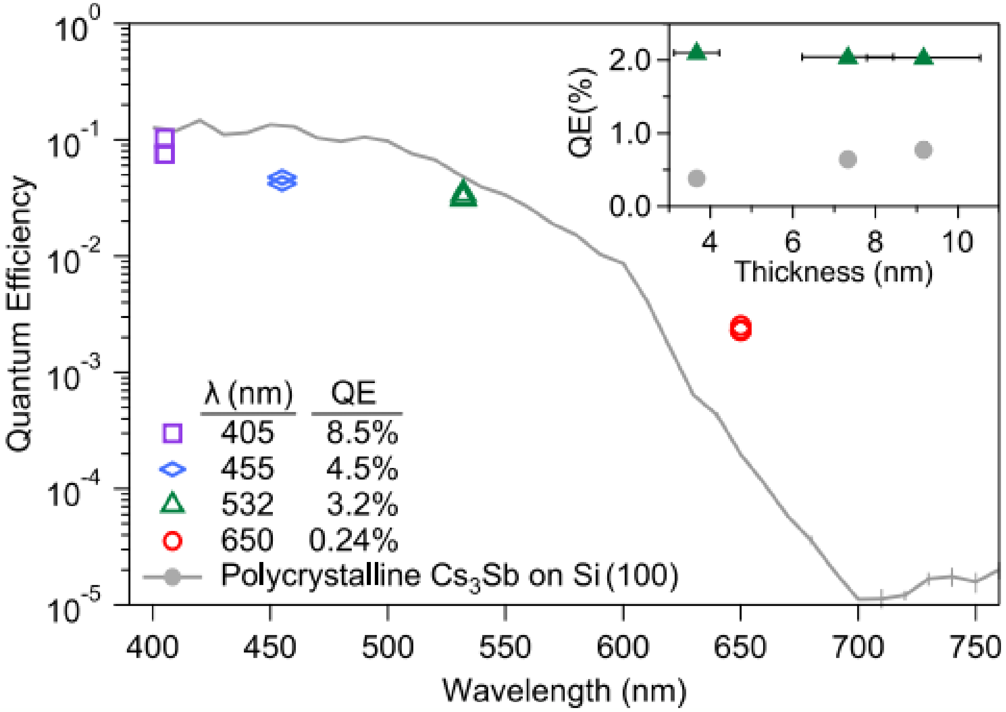
- Cs_3Sb grown via molecular beam epitaxy (MBE) on the lattice-matched 3C-SiC(100) substrate facilitates epitaxial growth.

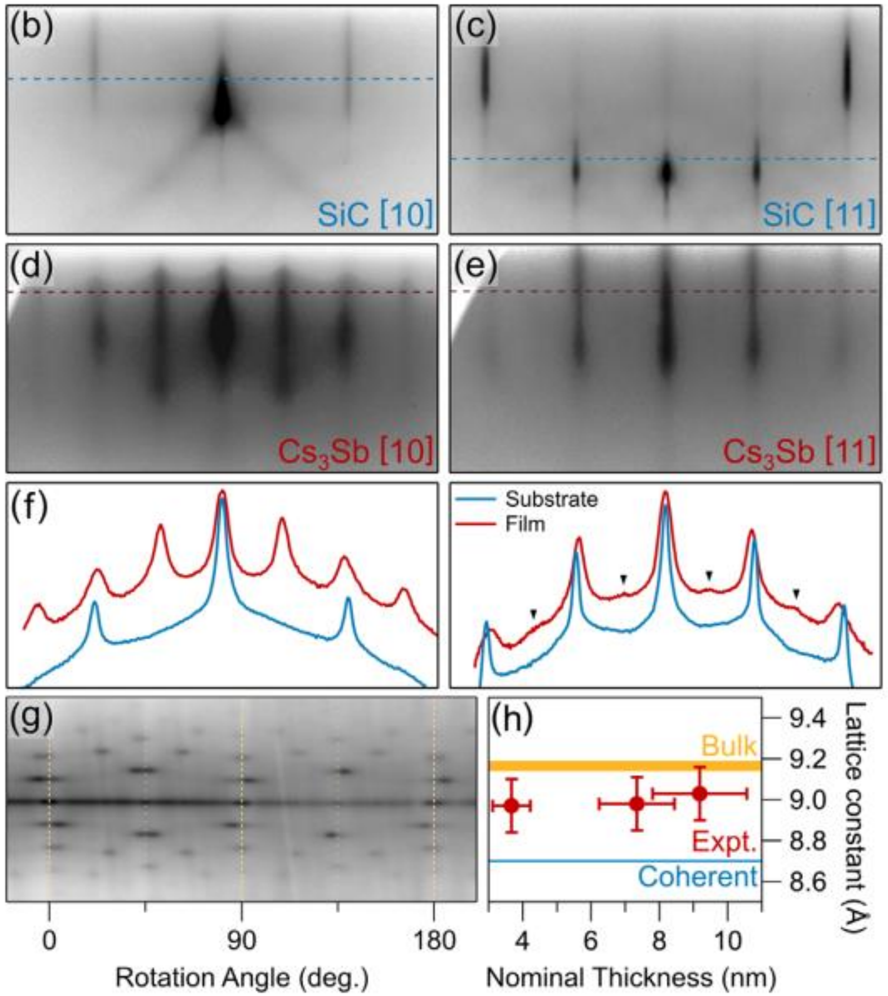


- C. T. Parzyck, A. Galdi, *et. Al.* Phys. Rev. Lett. 128, 114801 – Published 18 March 2022

Results on Cs_3Sb grown with MBE

- Cs_3Sb grown via molecular beam epitaxy (MBE) on the lattice-matched 3C-SiC(100) substrate facilitates epitaxial growth.
- Highly efficient in the ultra-thin limit (sub-10 nm). QE near 2% at 532 nm for ~2 nm thick film.





See John Smedley's talk in the Novel Concepts Session on Oct. 5!

- C. T. Parzyck, A. Galdi, *et. Al.* Phys. Rev. Lett. 128, 114801 – Published 18 March 2022

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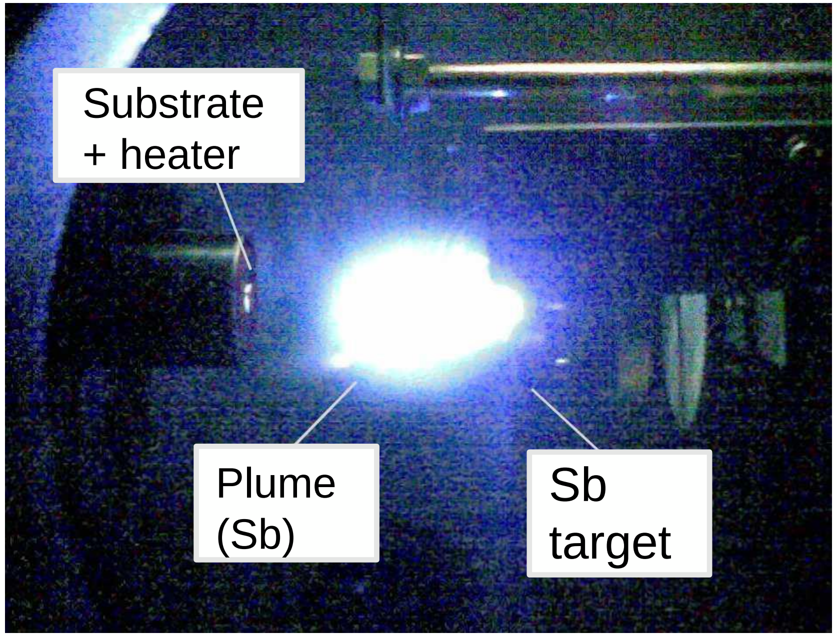
Photocathodes at NSLS-II

Beamline 4-ID at the National Synchrotron Light Source (NSLS-II):

- Option for growing with PLD or conventional thermal evaporation.
- Pulsed Laser Deposition (PLD)
  - 248 nm pulsed excimer laser vaporizes Sb target, condensing on substrate.
  - Consistent rep rate and laser fluence leads to extremely stable and controlled deposition rates.
  - Plumes of Sb or Sb_4?



Example PLD setup

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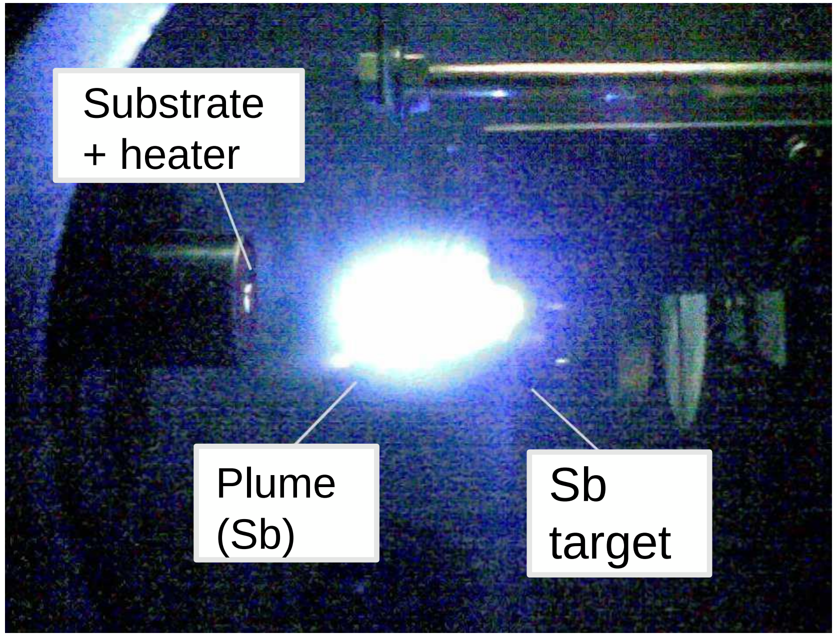
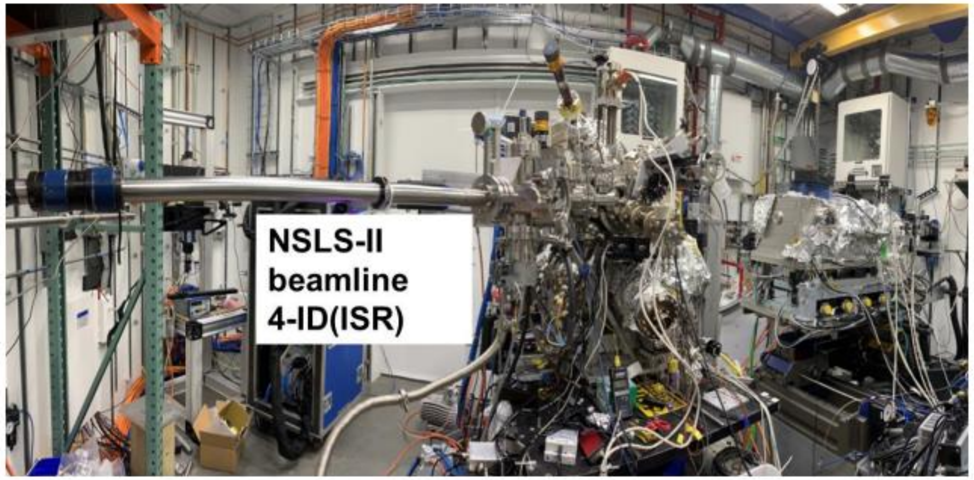


Example PLD setup

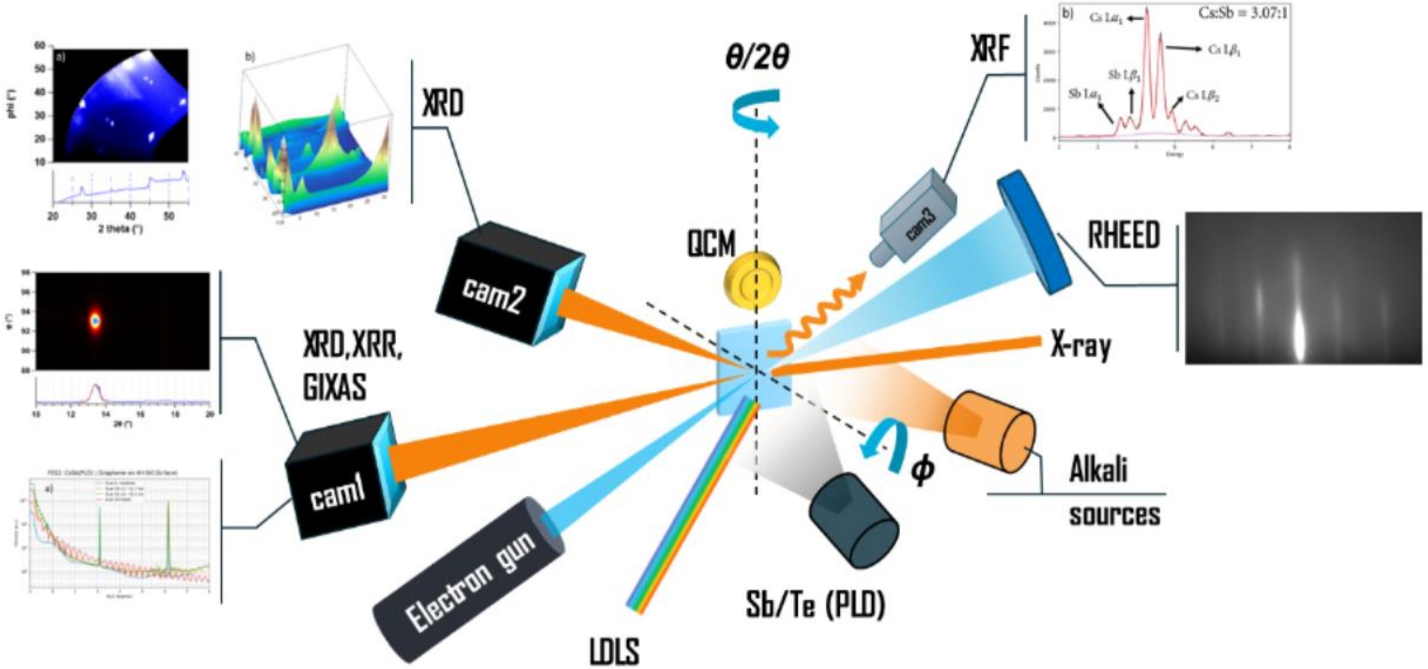
Characterization:

- X-ray diffraction (XRD) – bulk structure
- X-ray reflectivity (XRR) – thickness and rms roughness.
- X-ray fluorescence (XRF) – stoichiometry
- RHEED – surface structure
- Quantum efficiency



The experiment

- Our growth method:
  - Real-time thickness, stoichiometry, XRD measurements.



The experiment

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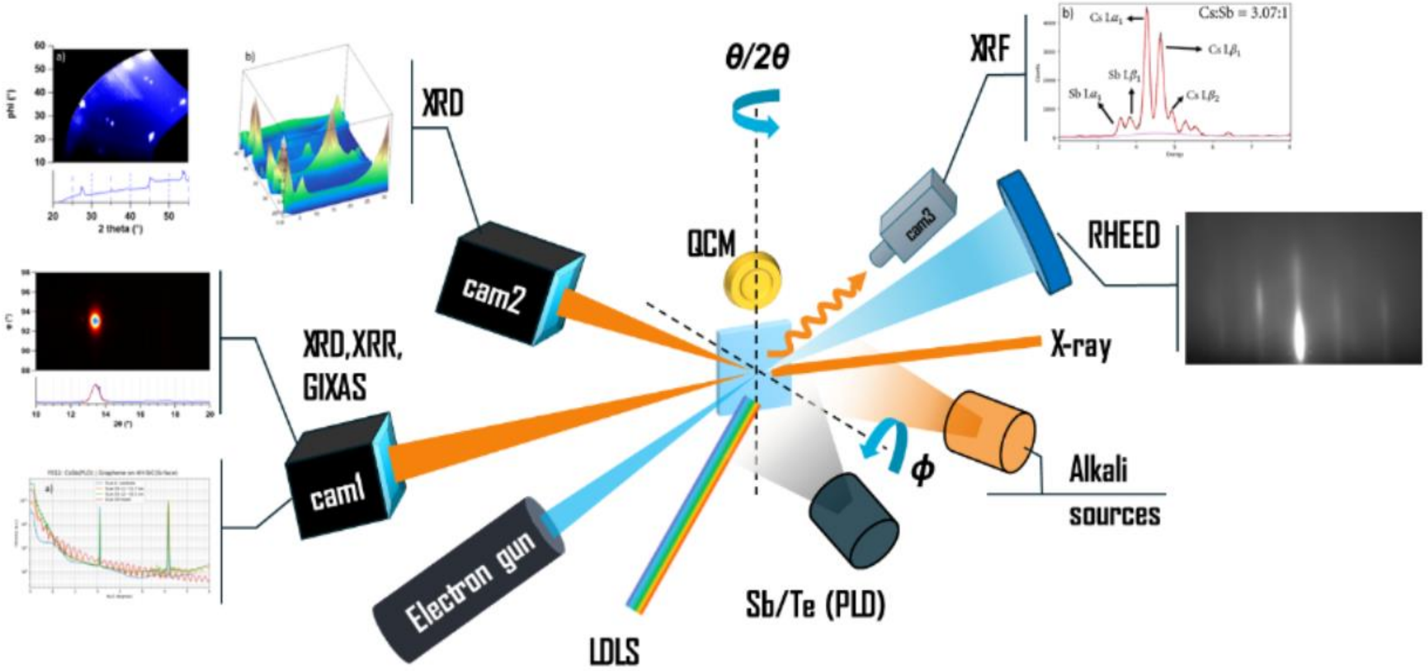
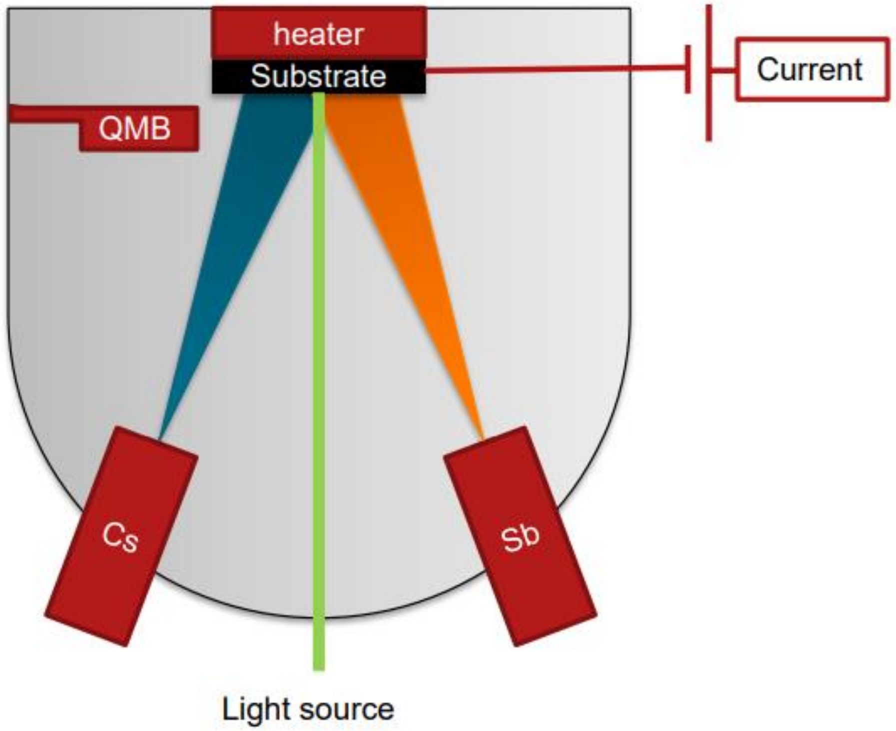
  - Real-time thickness, stoichiometry, XRD measurements.

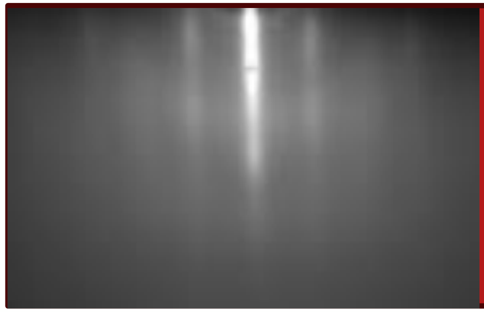
- Conventional photocathode growth:
  - Photocurrent oriented.
  - Maximize quantum efficiency.



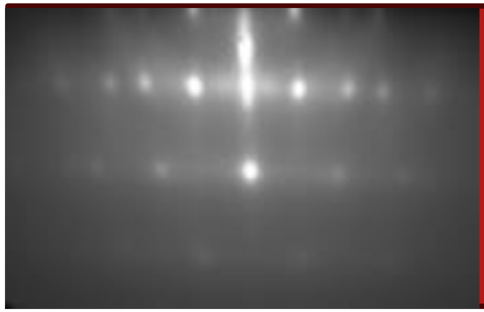


Flat, ordered Cs_3Sb on 3C-SiC(001)

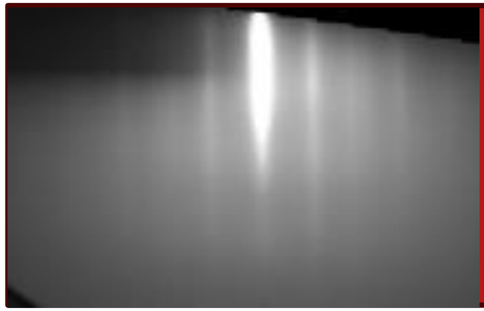
- RHEED patterns indicate flat and fiber-textured films.



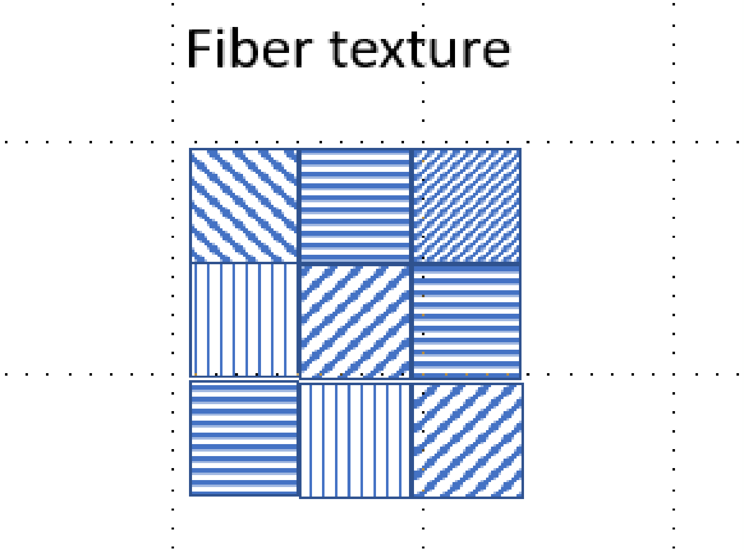
Deposition temperature: 90 C.
Grown using PLD.
Fiber textured film.
Thickness ~8 nm.



Deposition temperature: 80 C.
Grown using PLD.
Ordered film with 3D islands.
Thickness = 5.6 nm.



Deposition temperature: 85 C.
Grown using thermal evaporation.
Fiber textured film.
Thickness = 8.3 nm.



- Flat, thin, and ordered films with near percent level QE at 530 nm are easily grown with PLD.
- A Cs:Sb stoichiometry ratio of 3:1 is derived from XRF for ordered Cs_3Sb films.

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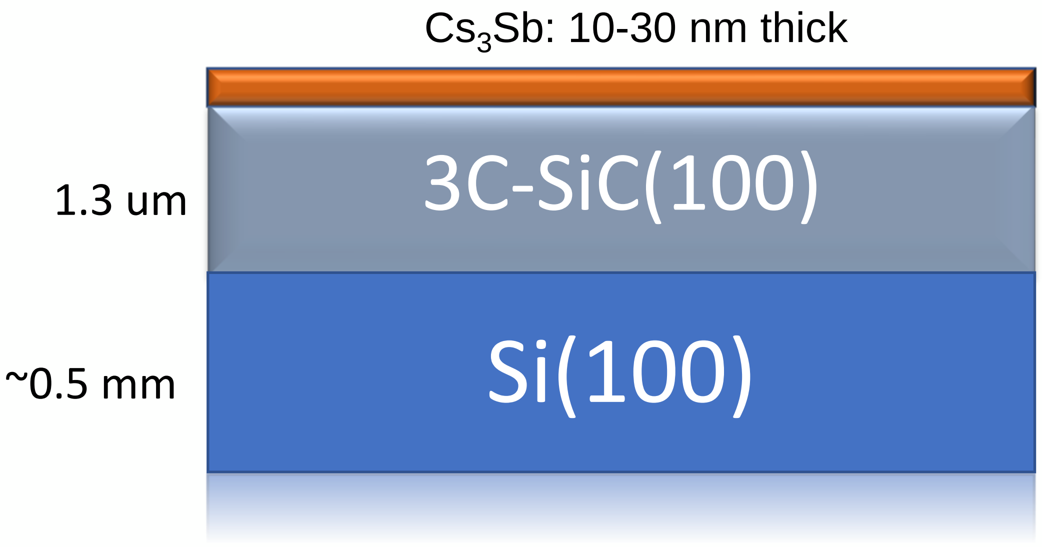
Optical interference in the cathode substrate



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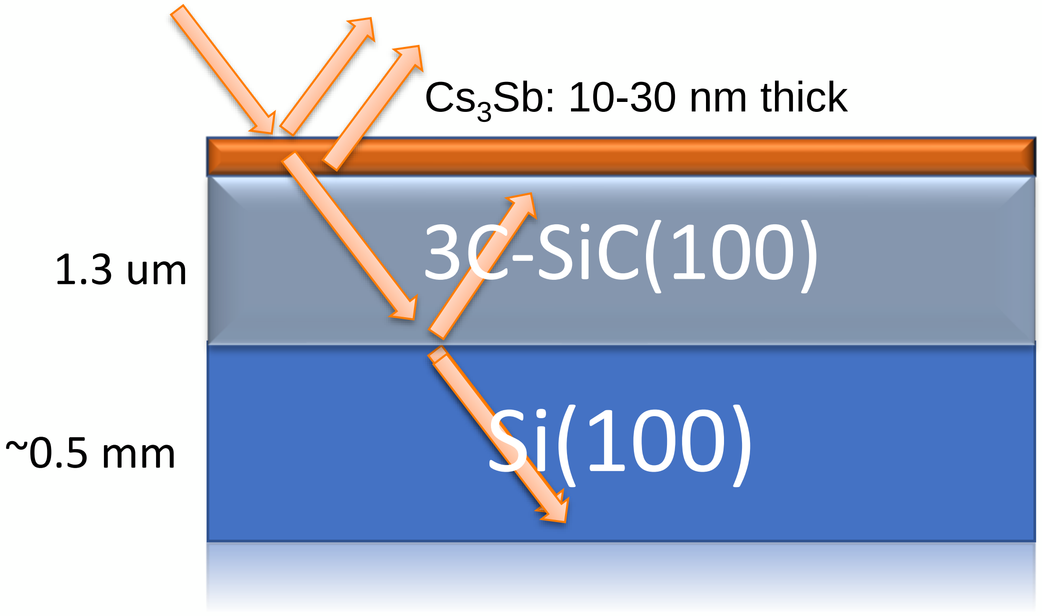
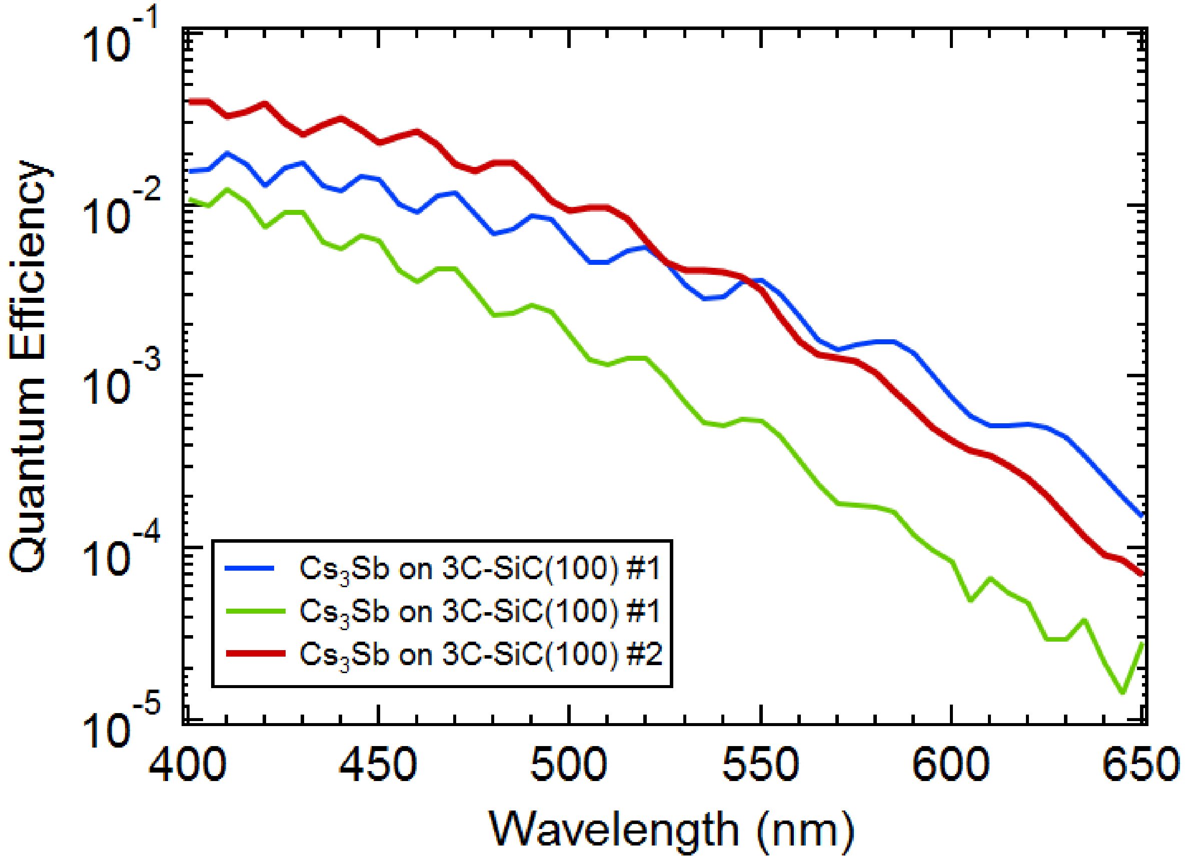


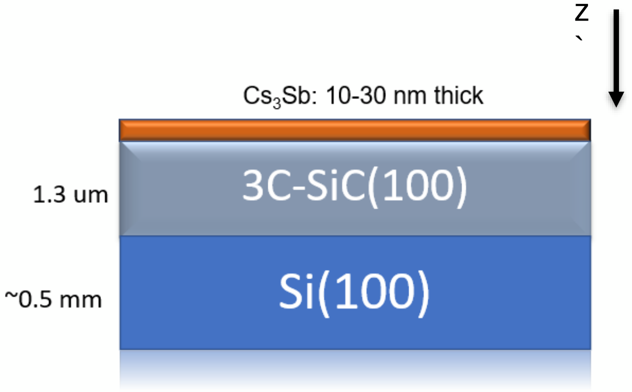


- Photocathode films are smooth and thin enough to measure effects from interference.

•**A. Alexander *et. al.* Enhanced photocathode performance through optimization of film thickness and substrate**
Journal of Vacuum Science & Technology B **35**, 022202 (2017); https://doi.org/10.1116/1.4976527

Optical interference in the cathode substrate

a.) 1.3 um SiC layer



<u>Electric field inside the cathode film</u>

$$E_y = E_0 e^{i\omega t}(c_f e^{i n_{PC} kz} + c_b e^{-i\tilde{n}_{PC} kz})$$

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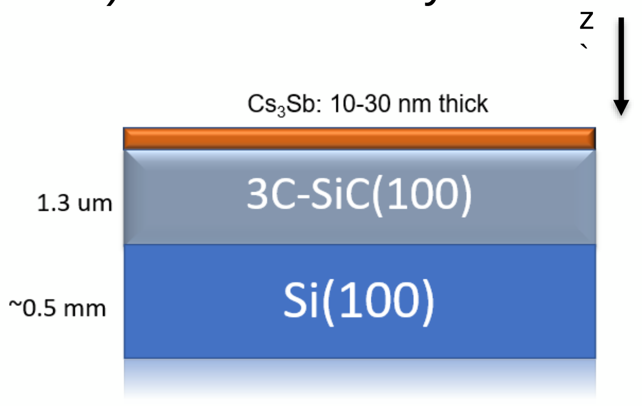

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<u>Calculate the Poynting vector</u>

$$S_z = \frac{1}{2} E_y \cdot \widetilde{H}_x$$

<u>Laser power absorbed in film</u>

$$P_{abs}(z) = \frac{d(Re(S_z))}{dz}$$

$$a(z, \lambda) = \frac{P_{abs}(z, \lambda)}{P_{in}(\lambda)}$$

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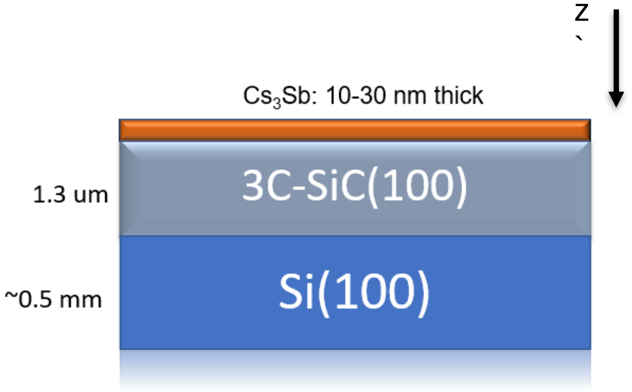

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Consider photoelectrons with energy greater than the workfunction. [2]

Profile of photon absorption into material.

$$QE = N \cdot \left(\frac{hc}{\lambda} - E_{vac}\right)^2 \int_0^h e^{-\frac{z}{\lambda_{esc}}} a(z,\lambda)dz$$

Probability of a photoexcited electron escaping material.

Where N is a pre-factor, h is the cathode thickness, E_{vac} is the vacuum energy, and λ_{esc} is the escape depth of the electron.

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- David H. Dowell and John F. Schmerge
Phys. Rev. ST Accel. Beams **12**, 074201 – Published 27 July 2009; Erratum Phys. Rev. ST Accel. Beams **12**, 119901 (2009)

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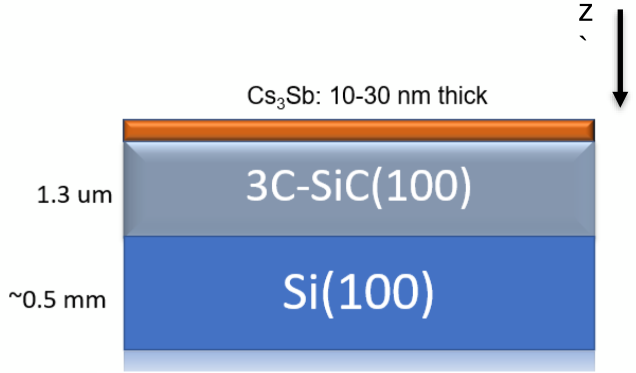

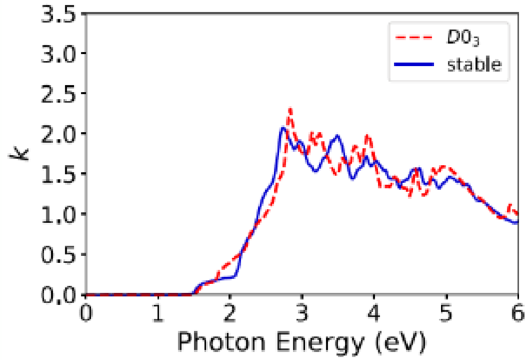

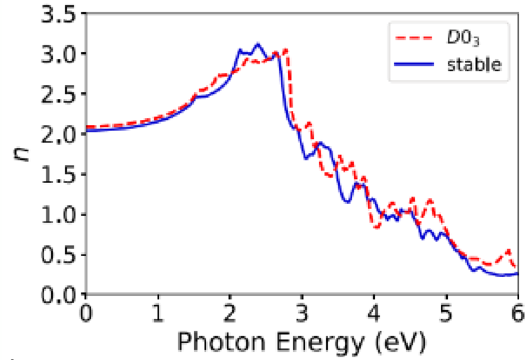

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Probability of a photoexcited electron escaping material.

Where N is a pre-factor, h is the cathode thickness, E_{vac} is the vacuum energy, and λ_{esc} is the escape depth of the electron.

- - Index of refraction data for Cs_3Sb obtained from DFT calculations.
- - Optical constants data for Si and SiC obtained from literature sources at https://refractiveindex.info/.
- Model could be improved with DFT density of states and detailed quantum mechanical descriptions.

See Dimitre Dimitrov talk in Theory session today!

Optical interference in the cathode substrate

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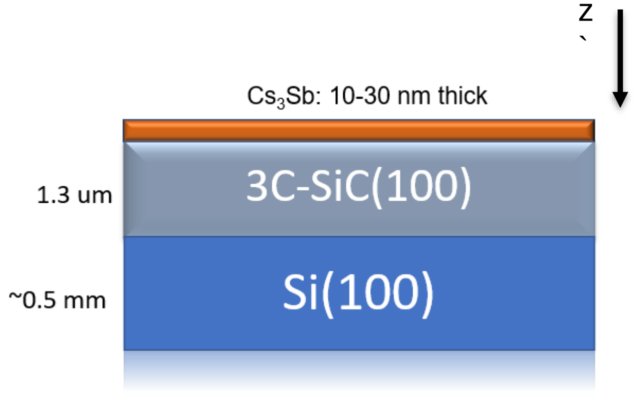
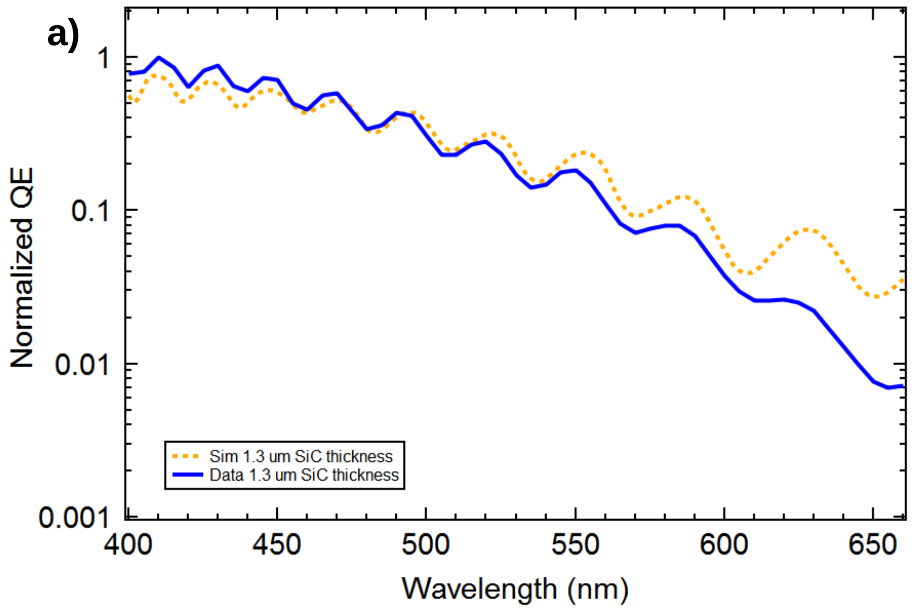
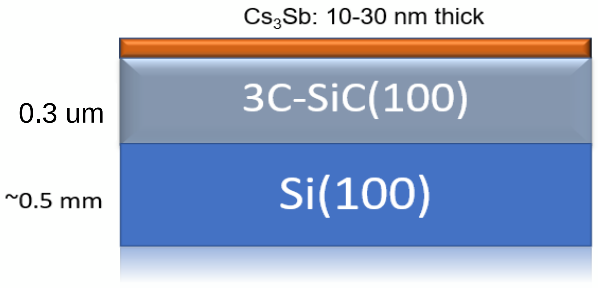
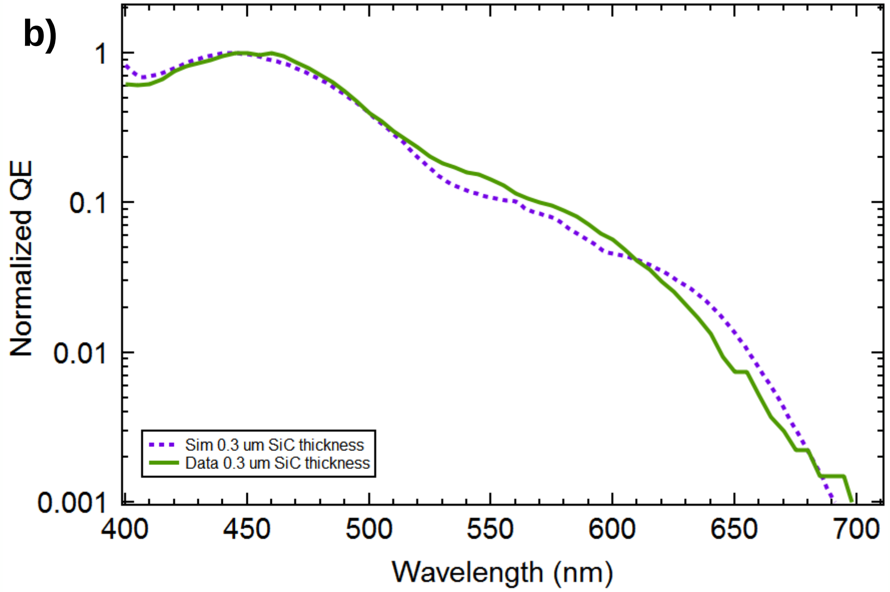




Fit results:
SiC thickess: 1.58 um.
Cs3Sb thickness: 35 nm.
e^- Escape depth: 60 nm.
Vacuum energy: 1.9 eV

b.) 0.3 um SiC layer





Fit results:
SiC thickess: 0.29 um.
Cs3Sb thickness: 23 nm.
e^- Escape depth: 100 nm.
Vacuum energy: 1.9 eV

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An alternate stoichiometry: Cs_1Sb_1

- While Cs_1Sb_1 is not epitaxial with the 3C-SiC substrate, smooth, fiber textured films are readily grown in wide range of substrate temperatures and deposition rates.

Azimuthally independent streaks in RHEED → fiber-textured surface

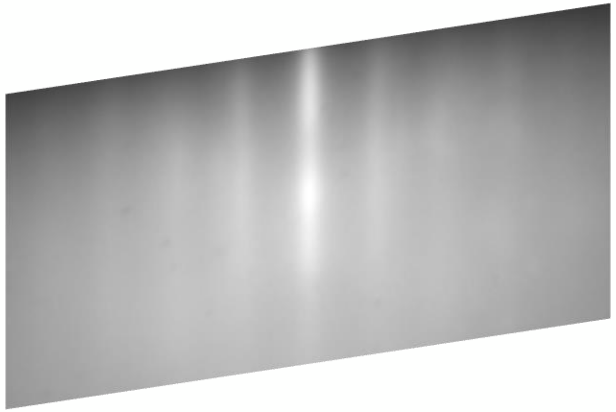
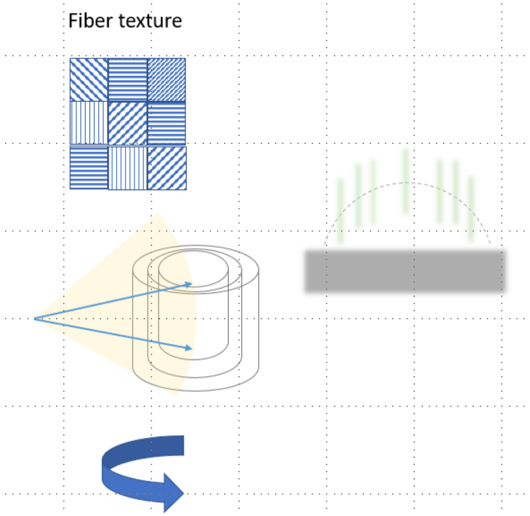
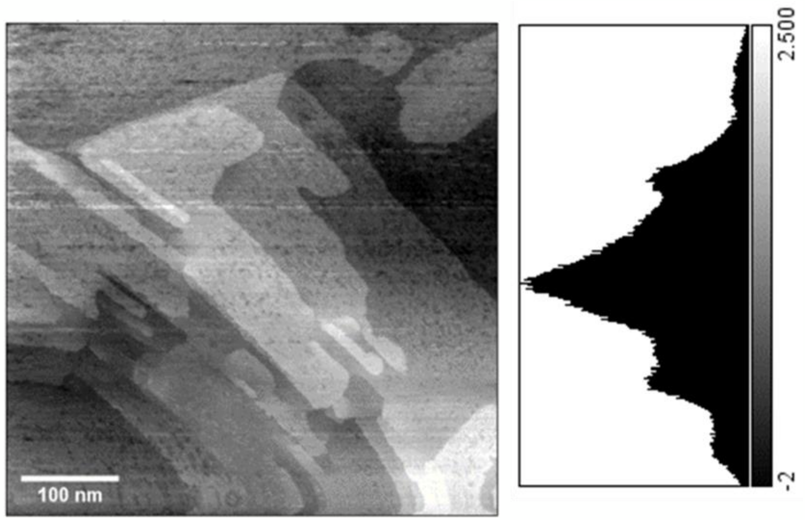


Image band pass filtered for contrast



Sample grown at:

- Substrate temp: 160 C.
- Thickness ~18 nm.
- CsSb samples were grown with both MBE and PLD.



Scanning Tunneling Microscopy (STM)

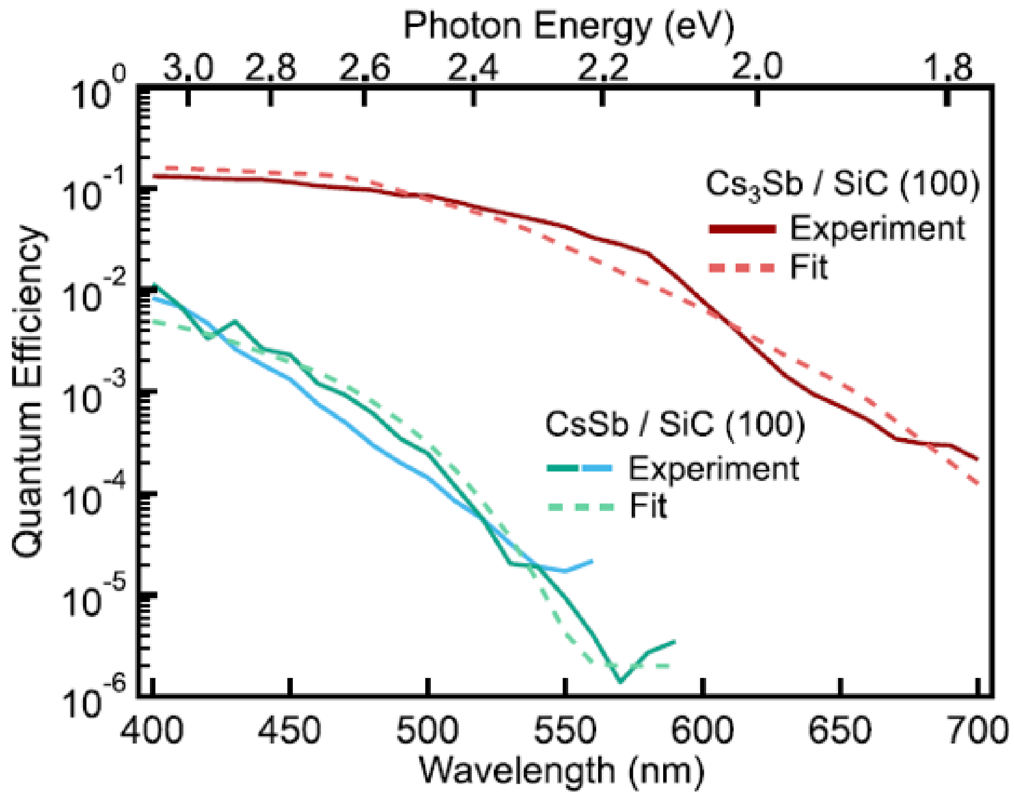
- Surface roughness ~0.6 nm.

-Multimodal histogram indicates flat steps along surface.

STM results courtesy of Hines Lab at Cornell.

An alternate stoichiometry: Cs_1Sb_1

- A visible photocathode with threshold near 570 nm and percent level QE at 400 nm.



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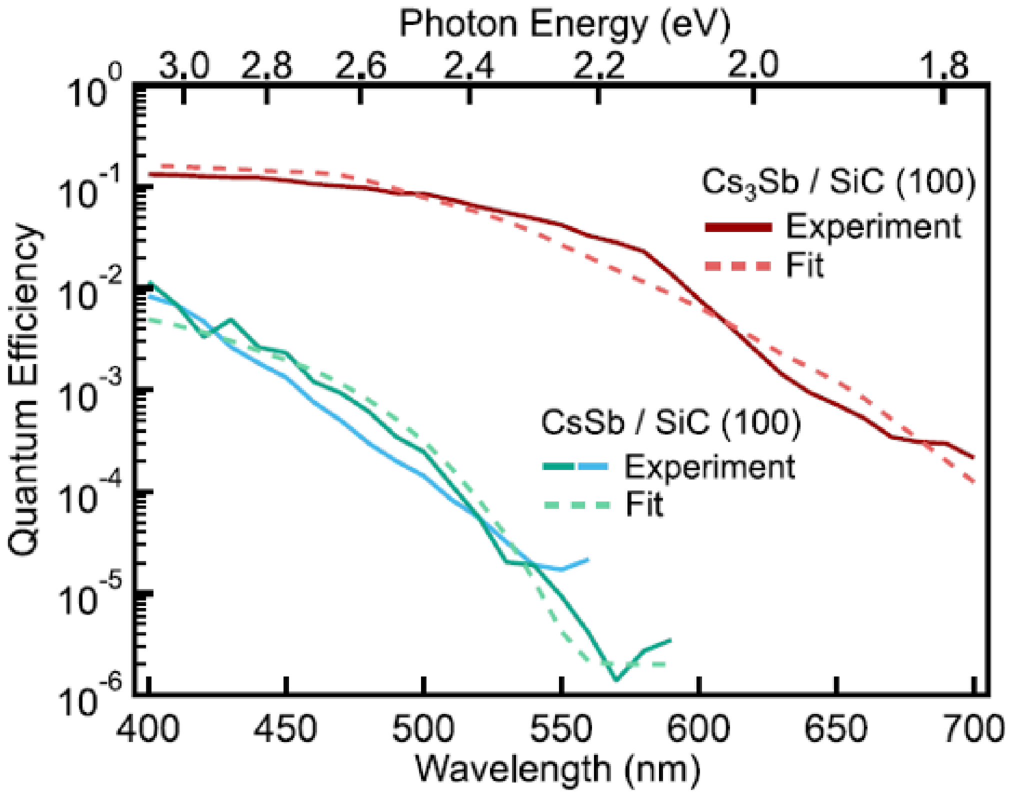
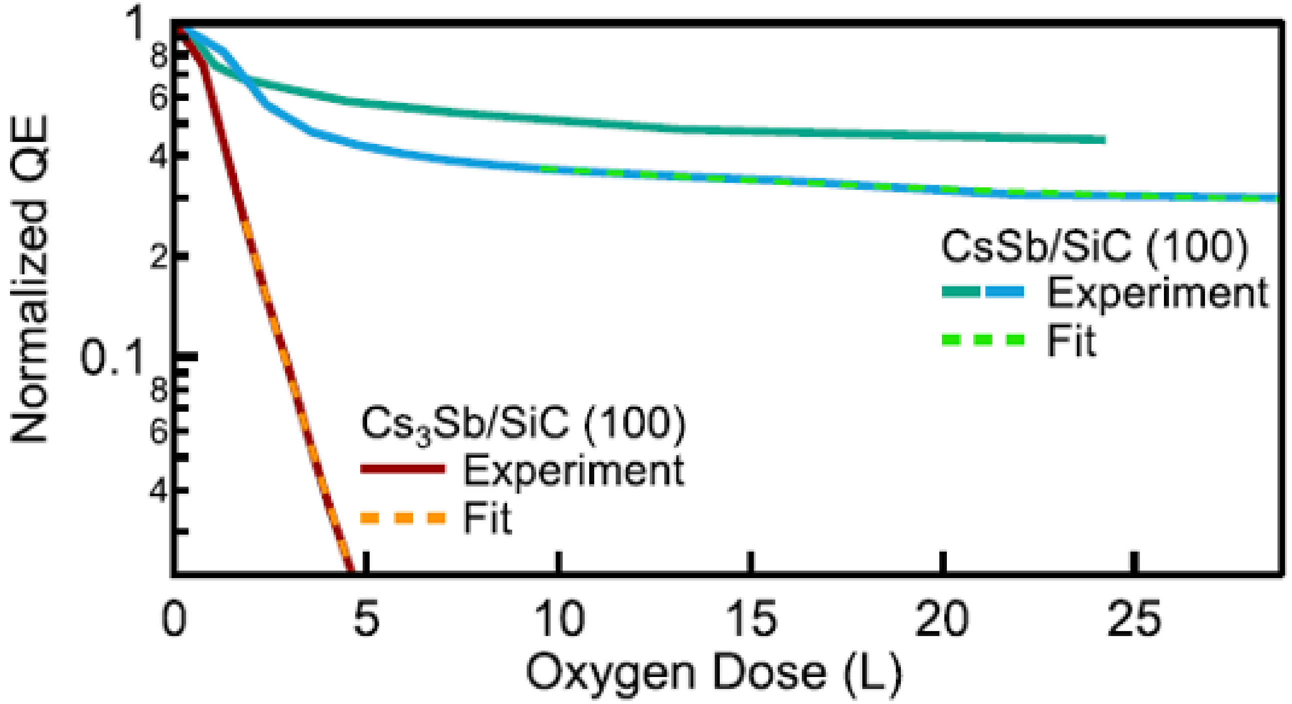


- Highly resistant to oxidation.
- Survives over an order of magnitude times longer than Cs_3Sb at an O_2 partial pressure of 5×10^{-8} Torr.



- Photoemission threshold lies between Cs_2Te and other alkali antimonides.

- C.T. Parzyck, C.A. Pennington et al, “Atomically smooth films of CsSb: a chemically robust visible light photocathode” arXiv:2305.19553

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Summary and Conclusions

- Flat, ordered, Cs_3Sb films with high quantum efficiency can be readily grown on commercially available, single crystal substrates (3C-SiC).

- Engineering quantum efficiency by exploiting optical interference effects in the cathode-substrate multilayer.

- Cs_1Sb_1 is a flat, oxygen-resistant photocathode with photoemission response at key laser wavelengths of 400 nm and 570 nm.

Future Directions:

- Investigate the relationship between epitaxy and intrinsic emittance.
- Explore epitaxial growth with bi-alkali antimonides (NaK_2Sb, K_2CsSb, etc.).
- Epitaxy opens the door to engineering photocathodes at the level of atomic layers to enhance brightness.

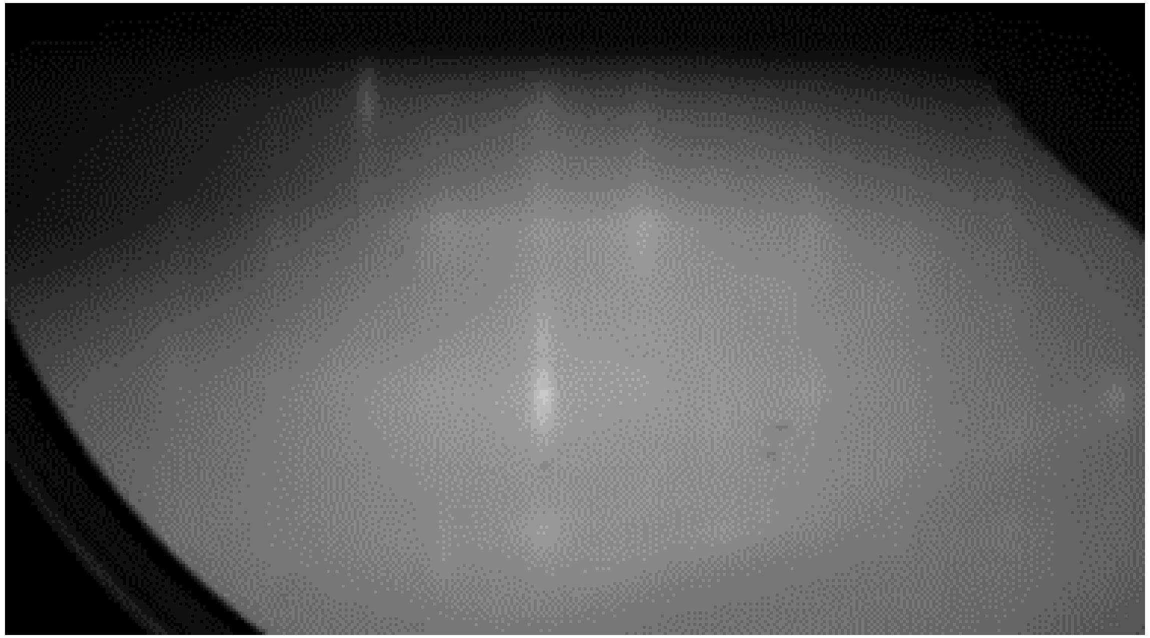
Thank you for listening!

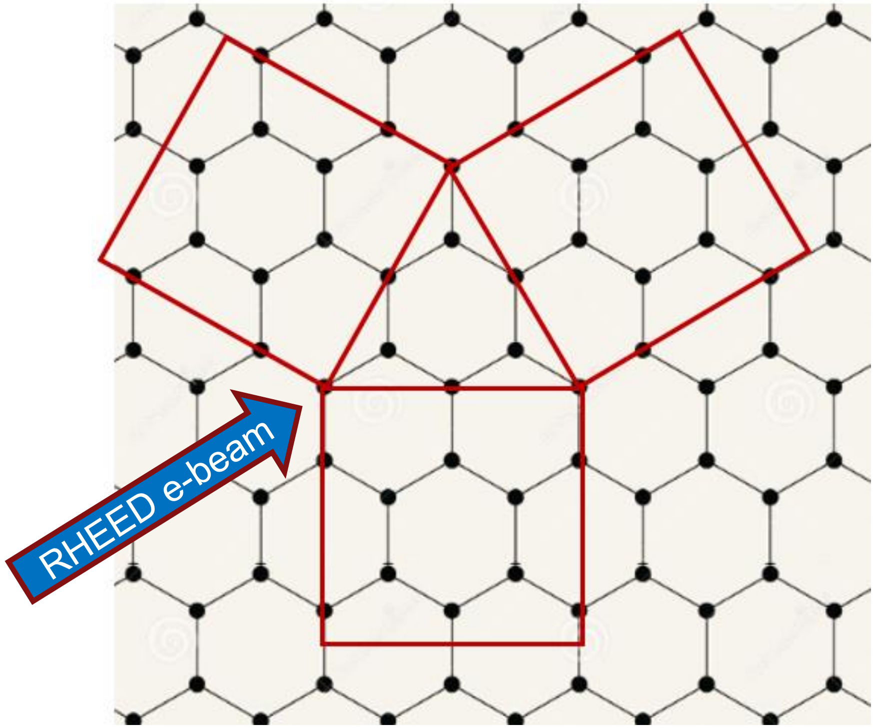
- Thanks to all the collaborators!
- Any questions?

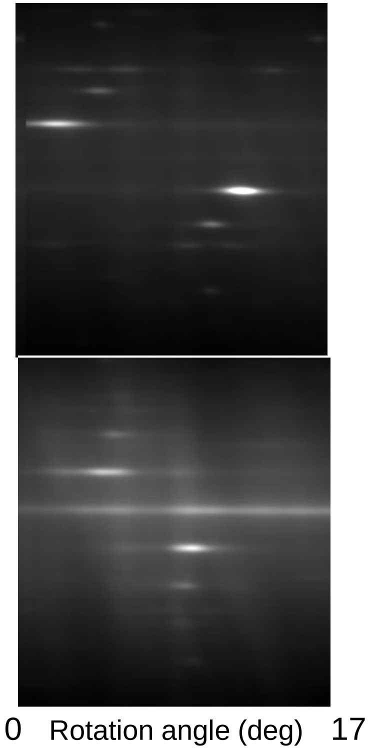
Bonus Slides

Epitaxial K_2CsSb on new substrate

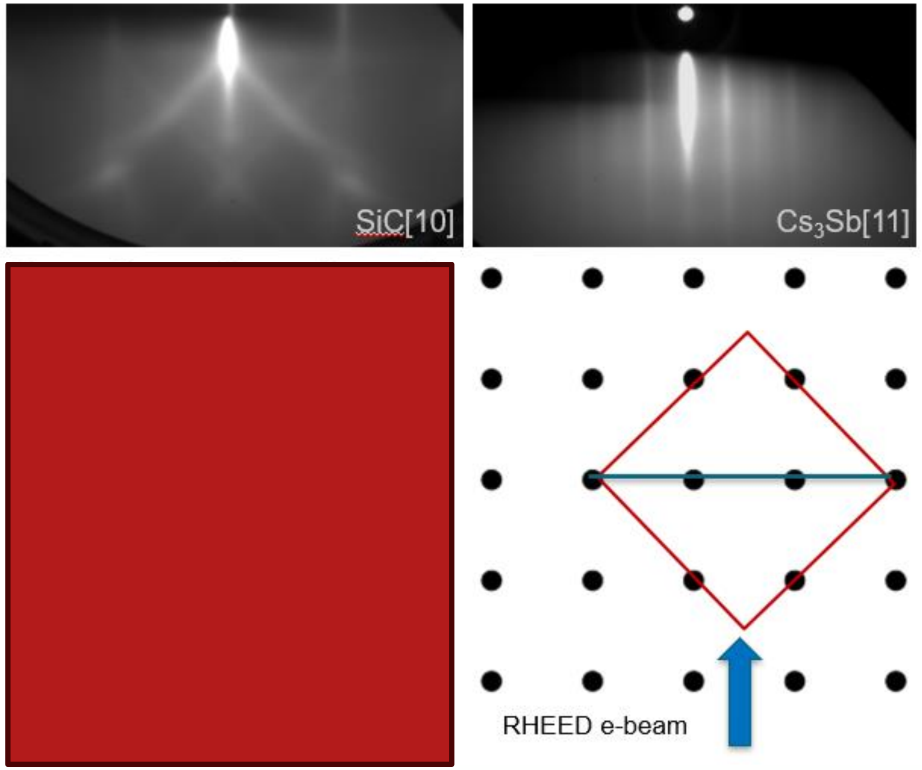
Growth conditions
Substrate temp: 85-90 C.
Sb deposition by PLD.
New substrate.
Ternary evaporation of K, Cs, Sb.

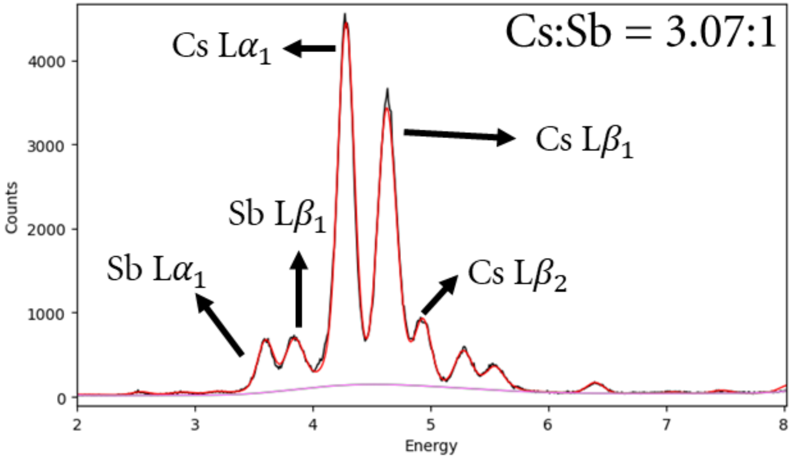






- K_2CsSb lattice constant: ~8.61 Angs
- K_2CsSb grows with epitaxial domains on new substrate.





Representative Cs_3Sb on 3C-SiC(100) grown with PLD.
Substrate temperature held at 90 degrees.