Epitaxial Growth of Cesium Potassium Antimonide Photocathode

Presenter(s): Kali Prasanna Mondal, Collider-Accelerator Department

Contributors: Mengjia Gaowei (Supervisor), John Smedley, Jared Maxson, Chad Pennington, Elena Maria Echeverria Mora, Kenneth Evans-Lutterodt, Raul Acevedo-Esteves, Siddharth Karkare, Priyadarshini Bhattacharyya, Pallavi Saha, Jean Jordan-Sweet, Guido Stam, Molen S.J. Van der, Thomas Juffmann, Rudolf Tromp, John Walsh, Rudy Begay

Date: 10/03/2023

Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ Experimental Details
- ❑ PLD: epitaxial growth
    - ❑ RHEED, XRD, XRR, GISAXS, XRF results
    - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
    - ❑ RHEED results
    - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

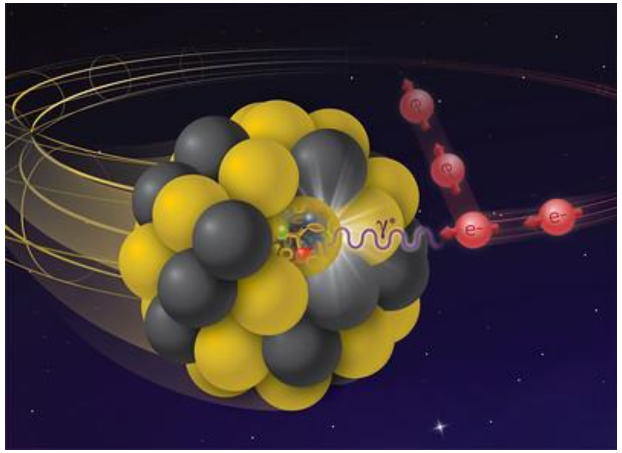
Brookhaven National Laboratory

Outline

- ❑ **Introduction: Photocathode and epitaxial growth**
- ❑ Experimental Details
- ❑ PLD: epitaxial growth
  - ❑ RHEED, XRD, XRR, GISAXS, XRF results
  - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
  - ❑ RHEED results
  - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

Unpolarized photocathodes for EIC electron cooling

- To maintain a luminosity of L= $10^{34} cm^{-2} s^{-1}$ in the Electron Ion Collider (EIC) during long collision runs, it is desirable to cool the hadron beams in order to balance emittance growth rates due to intrabeam scattering (IBS)
- Electron cooling is a promising technique to improve the luminosity of high current beams and has been demonstrated to cool proton beams



Photocathode

- Cathode is origin of the generated electrons, which could be a metal, a semiconductor.
- Photocathode: Convert photon to electron (photoelectric effect), electron emits in vacuum.
- Photoemission offers advantages, for example that electron bunches can be time structured.

Electron beam required for e-cooling in EIC

- High average current (> 100 mA)
- High bunch charge (1nC)
- Long lifetime (> 1 week)
- Low emittance

- Fast development of the particle accelerator devices and the strong desire to achieve higher bunch charges and currents leads to the usage of semiconductor photocathodes such as alkali antimonide (K_2CsSb), Cesium antimonide (Cs-Sb), cesium telluride (Cs_2Te) or gallium arsenide (GaAs).

Brookhaven National Laboratory

Epitaxial growth of Photocathode

- Epitaxy: The name epitaxy has Greek roots: “epi” means “on” and “taxis” means “in ordered manner”. Epitaxy refers to the growth of a single crystalline film on a single crystalline substrate.
- Epitaxial Growth of Photocathode: Large or single crystal photocathode generate low mean transverse energy due to smooth surface and low grain boundary scattering rate.
- Epitaxial growth with lattice matching substrate is essential to make large/single crystal photocathode.

Our goal

- Our main goal is to grow epitaxial semiconductor photocathode thin film.
- And achieve high quantum efficiency with epitaxial photocathode.

Introduction

- Epitaxial growth of Cesium Potassium Antimonide (K_2CsSb), deposited on different substrates: New substrate, 4H-SiC.
- Epitaxial growth: Compare PLD and thermal-assisted growths.

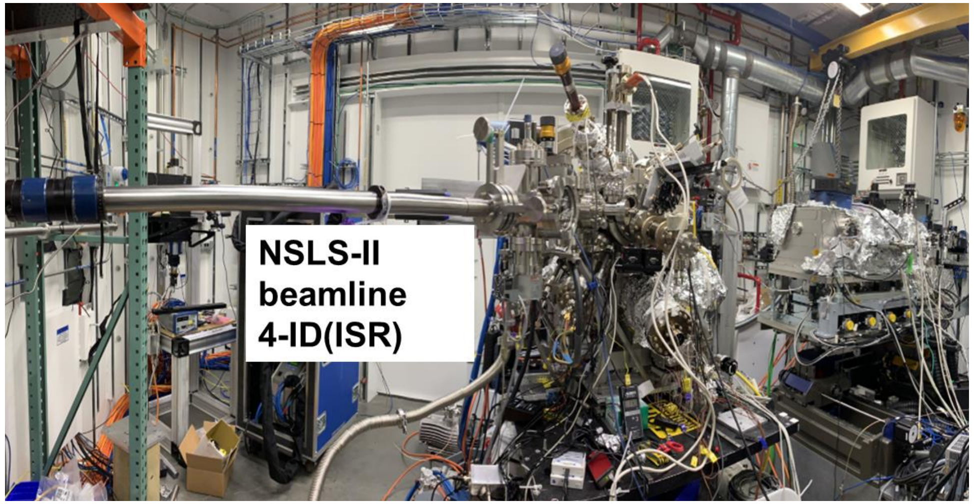
Brookhaven National Laboratory

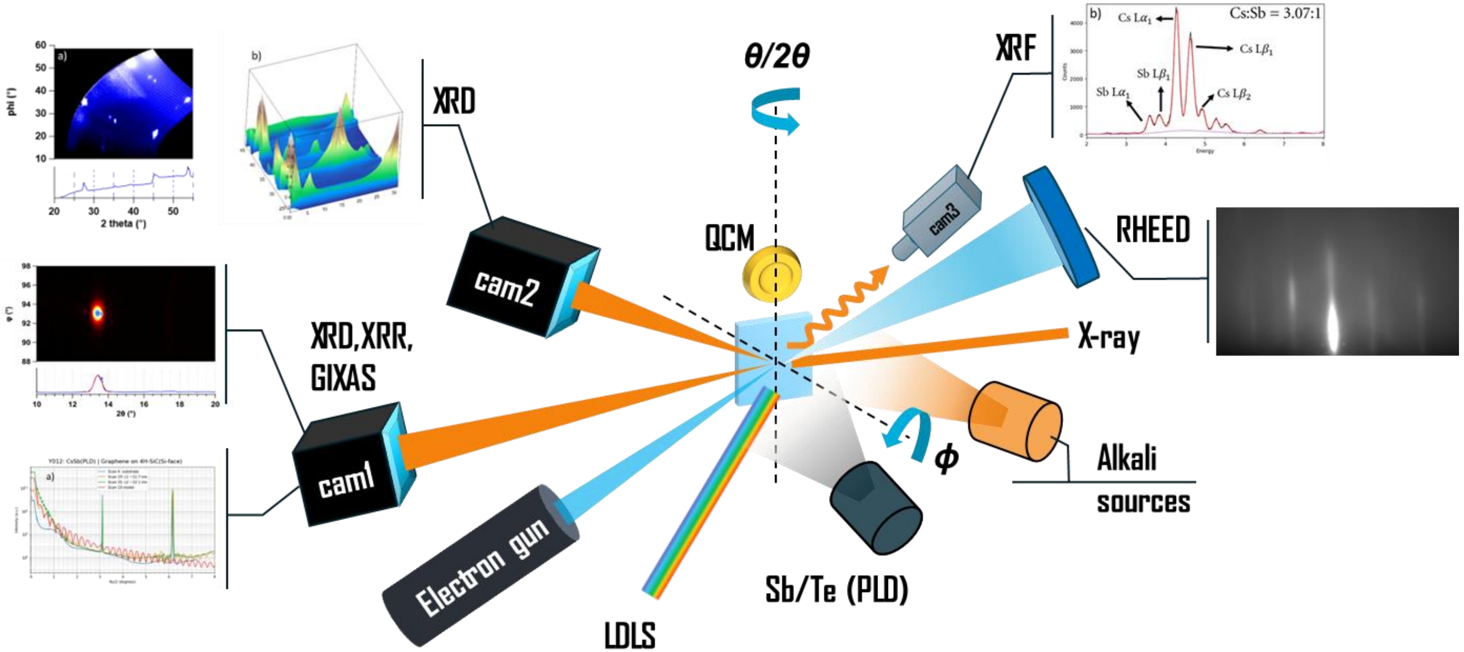
Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ **Experimental Details**
- ❑ PLD: epitaxial growth
  - ❑ RHEED, XRD, XRR, GISAXS, XRF results
  - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
  - ❑ RHEED results
  - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

Brookhaven National Laboratory

The BNL UHV photocathode growth system at beamline 4-ID, Integrated In situ and Resonant Hard X-ray Studies (ISR), NSLS-II for in situ and real time x-ray characterization. schematic of X-ray techniques (XRR, XRD, GISAXS, XRF) used to characterize the photocathode properties.





Evaporators:

- ❑ Thermal Sb/Te
- ❑ Alkali metals
- ❑ PLD Sb/Te

- ❑ K and Cs deposited using effusion cell.
- ❑ Sb deposited with both PLD and thermal growth.

Characterization:

- ❑ Reflection high energy electron diffraction (RHEED): epitaxial growth and crystalline structural details.
- ❑ X-ray diffraction (XRD): crystalline structure.
- ❑ X-ray reflectivity (XRR): thin film thickness, roughness, electron density.
- ❑ Grazing incidence small angle x-ray scattering (GISAXS): Structural details both from surface and interface.
- ❑ X-ray fluorescence (XRF): Stoichiometry of Photocathode
- ❑ Quantum efficiency (QE) measurement: QE of photocathodic thin film.
- ❑ Quartz crystal microbalance (QCM): thin film thickness

Epitaxial Growth of K_2CsSb

PLD and thermal evaporation assisted epitaxial growth
at 4ID, NSLS-II, BNL

Brookhaven National Laboratory

Sample 1	Sample 2	Sample 3
K_2CsSb	K_2CsSb	K_2CsSb
New Substrate	New Substrate	4H-SiC
❑ Sb deposited using PLD.	❑ Substrate is heat clean from Sample-1. ❑ Sb deposited using Thermal evaporation.	❑ Sb deposited using Thermal evaporation.

For all samples

- ❑ Cs and K were deposited using effusion cells.
- ❑ Layer 1: K_2CsSb on substrate, for structural study
- ❑ Layer 2: Growth of another layer of K_2CsSb on Layer 1
- ❑ Layer 3: Growth of photocathode high enough thickness to achieve high QE.

K_2CsSb (Layer 3)
K_2CsSb (Layer 2)
K_2CsSb (Layer 1)
Substrate

Brookhaven National Laboratory

Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ Experimental Details
- ❑ **PLD: epitaxial growth**
  - ❑ **RHEED, XRD, XRR, GISAXS, XRF results**
  - ❑ **Spectral response**
- ❑ Thermal: Epitaxial growth
  - ❑ RHEED results
  - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

Brookhaven National Laboratory

Sample 1	Sample 2	Sample 3
K_2CsSb	K_2CsSb	K_2CsSb
New Substrate	New Substrate	4H-SiC

Sample 1
- ❑ Sb deposited using PLD.

Sample 2
- ❑ Substrate is heat clean from Sample-1.
- ❑ Sb deposited using Thermal evaporation.

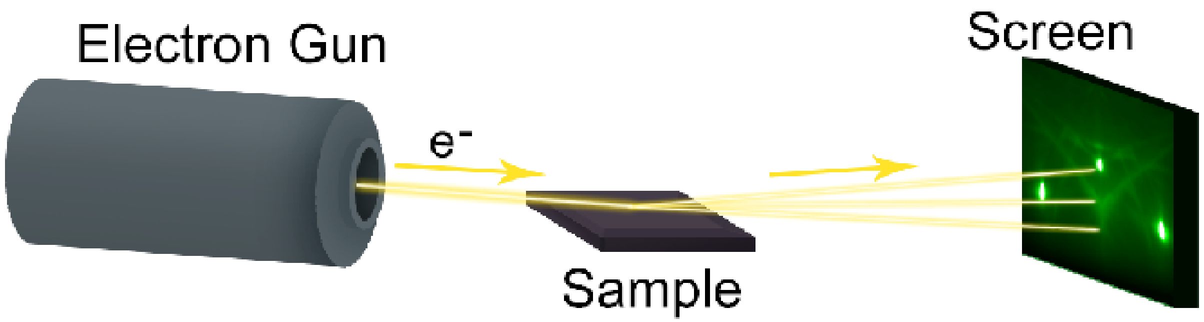
Sample 3
- ❑ Sb deposited using Thermal evaporation.

For all samples
- ❑ Cs and K were deposited using effusion cells.
- ❑ Layer 1: K_2CsSb on substrate, for structural study
- ❑ Layer 2: Growth of another layer of K_2CsSb on Layer 1
- ❑ Layer 3: Growth of photocathode high enough thickness to achieve high QE.

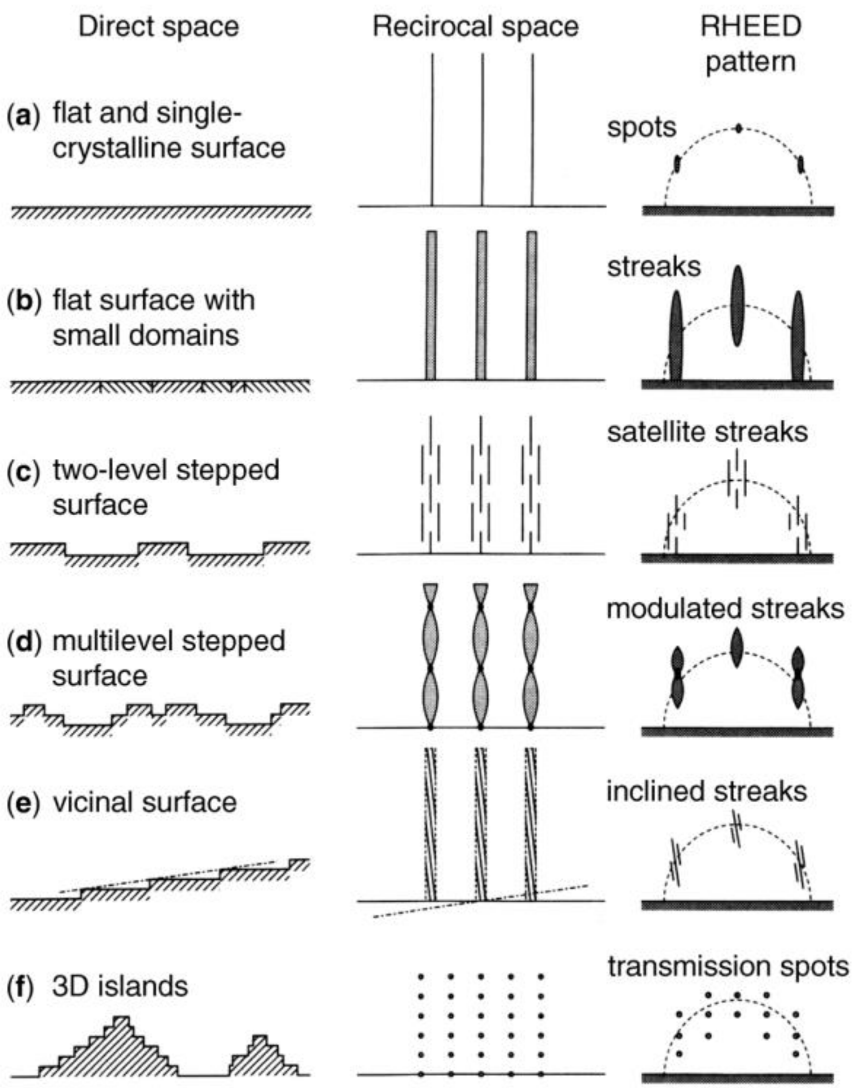
K_2CsSb (Layer 3)
K_2CsSb (Layer 2)
K_2CsSb (Layer 1)
Substrate

RHEED of Photocathode

- RHEED provides -surface symmetry, real space lattice spacing, crystalline degree of perfection
- Photocathodic thin film, K_2CsSb grown on 4H-SiC, new substrate.
- RHEED has been performed after the completion of each growth.
- Our goal is to
  - Study crystalline structure of photocathodic thin film after each growth
  - Also, study of angular dependence of crystalline structure with in-plane rotation
  - Find out lattice mismatch, strain of thin films



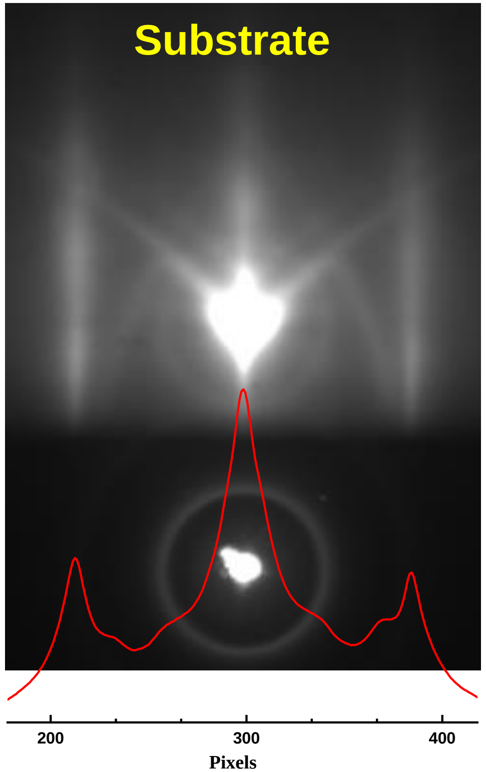
Schematic of RHEED [Derriche et al. 2019]

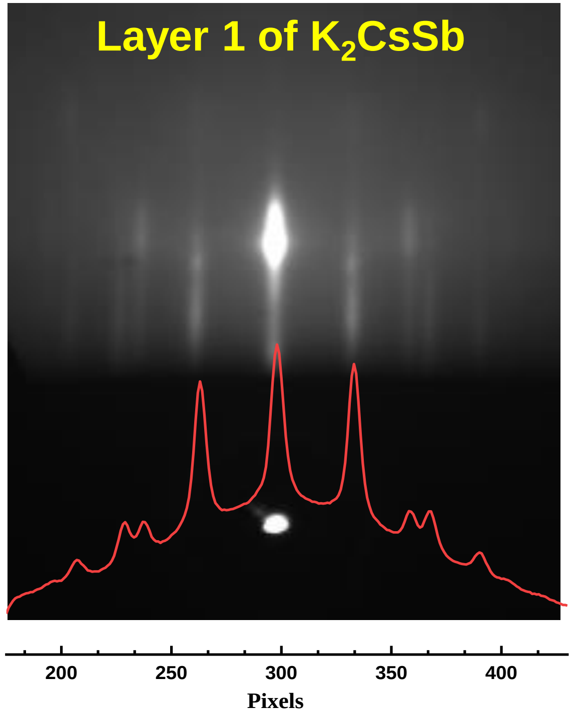


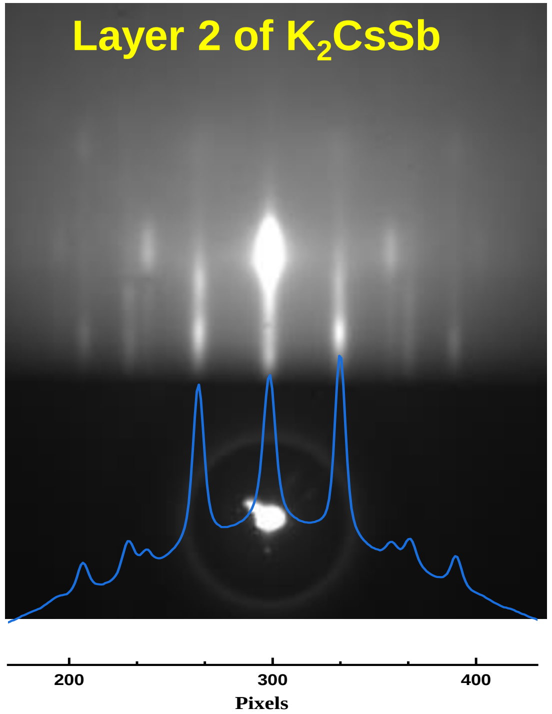
Schematics of various kinds of realistic surfaces, in real-space morphology, in reciprocal space, and their RHEED patterns [Hasegawa, 2012]

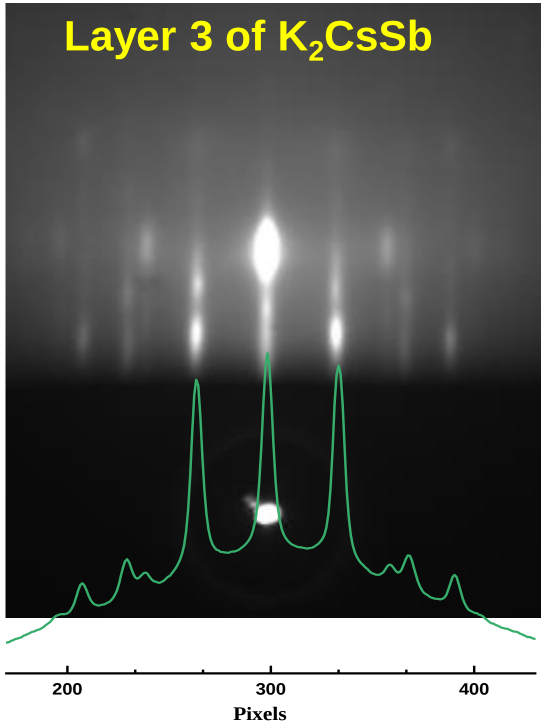
Brookhaven National Laboratory

RHEED of Sample 1: Sb deposited using PLD









XRF of K_2CsSb (Layer 3)

Cs/Sb: 1.29;
K/Sb:1.88

Brookhaven National Laboratory

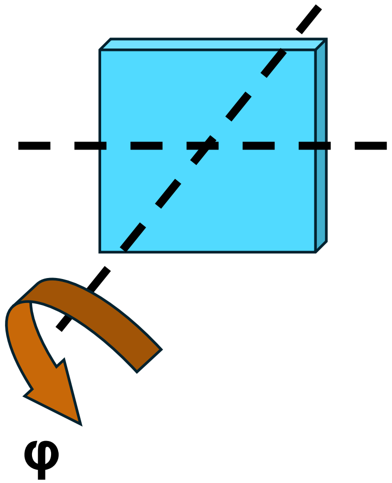
K_2CsSb/New Substrate (Layer 1)

Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	34.5	4.97	111
2nd	60	2.86	122
3rd	69	2.49	222
4th	91.5	1.87	133

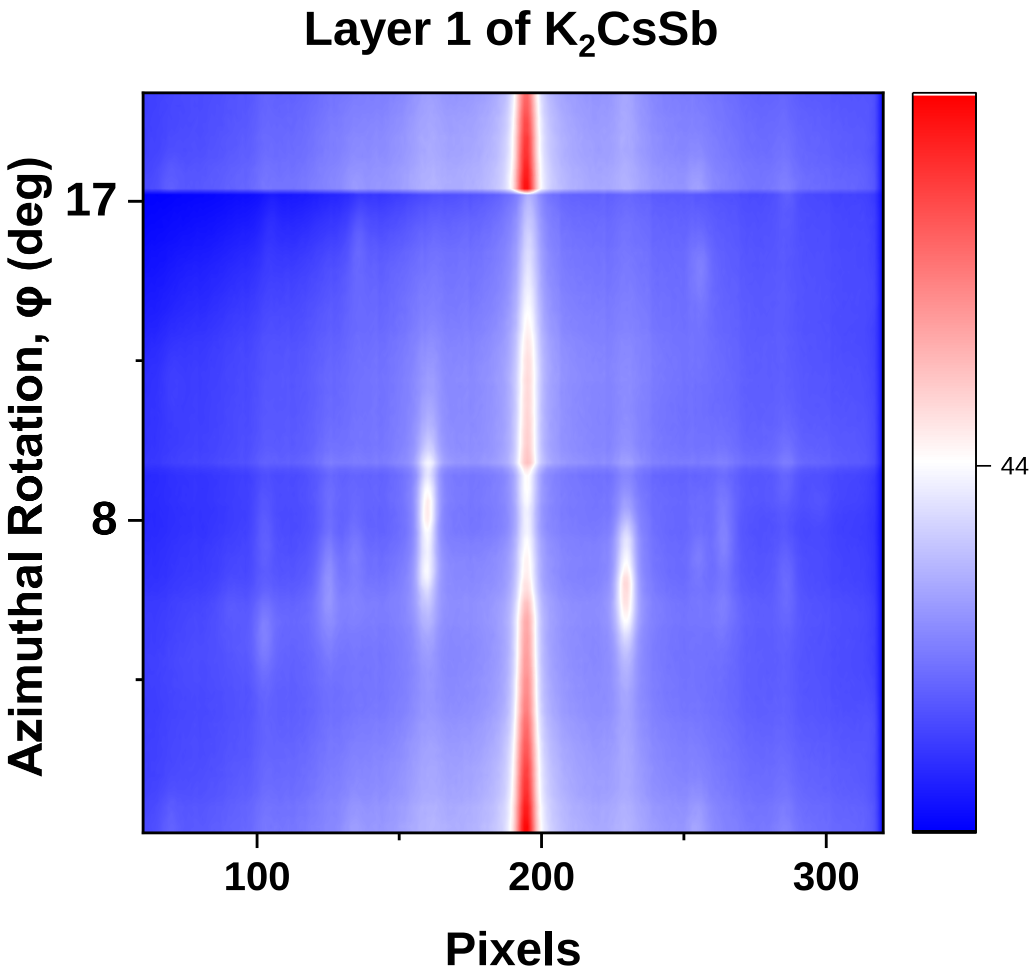
RHEED of K_2CsSb

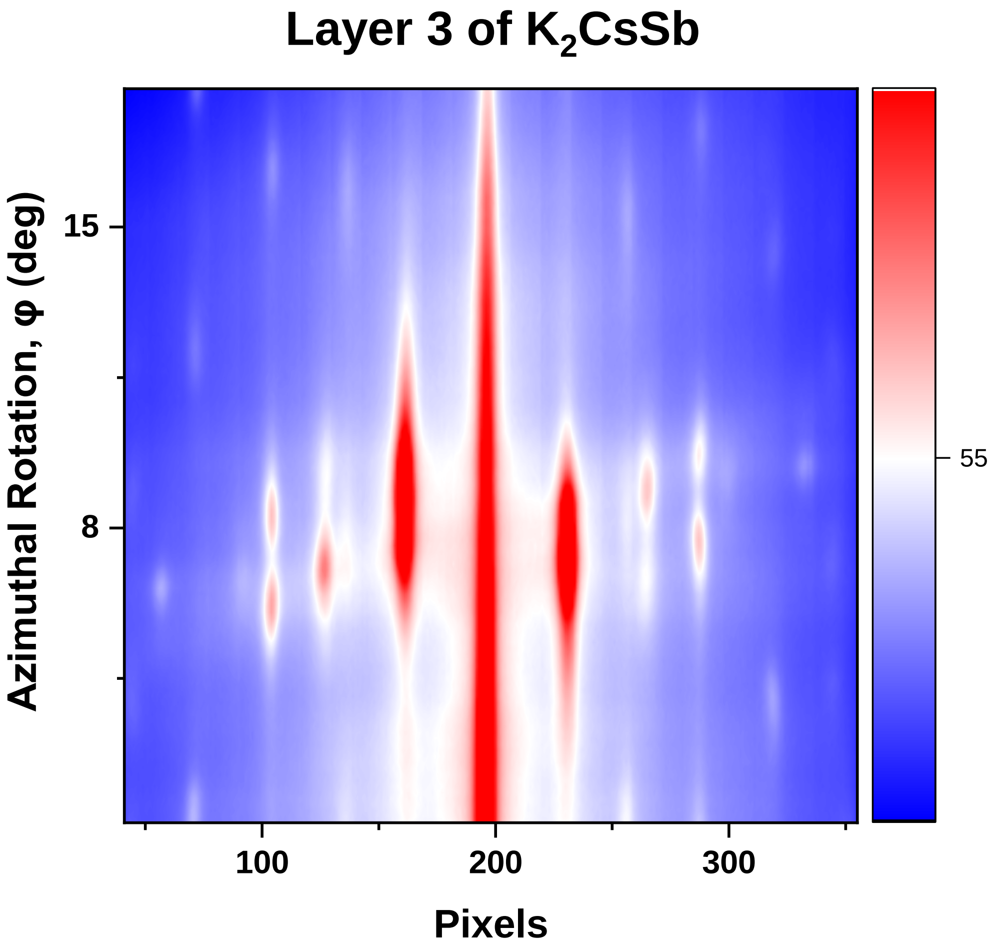
- RHEED image shows epitaxial growth.
- Streaks represent smooth surfaces with small domains.
- With increasing thickness roughness increases.

Azimuthal angular dependence of RHEED from Sample-1



Schematic of azimuthal sample rotation

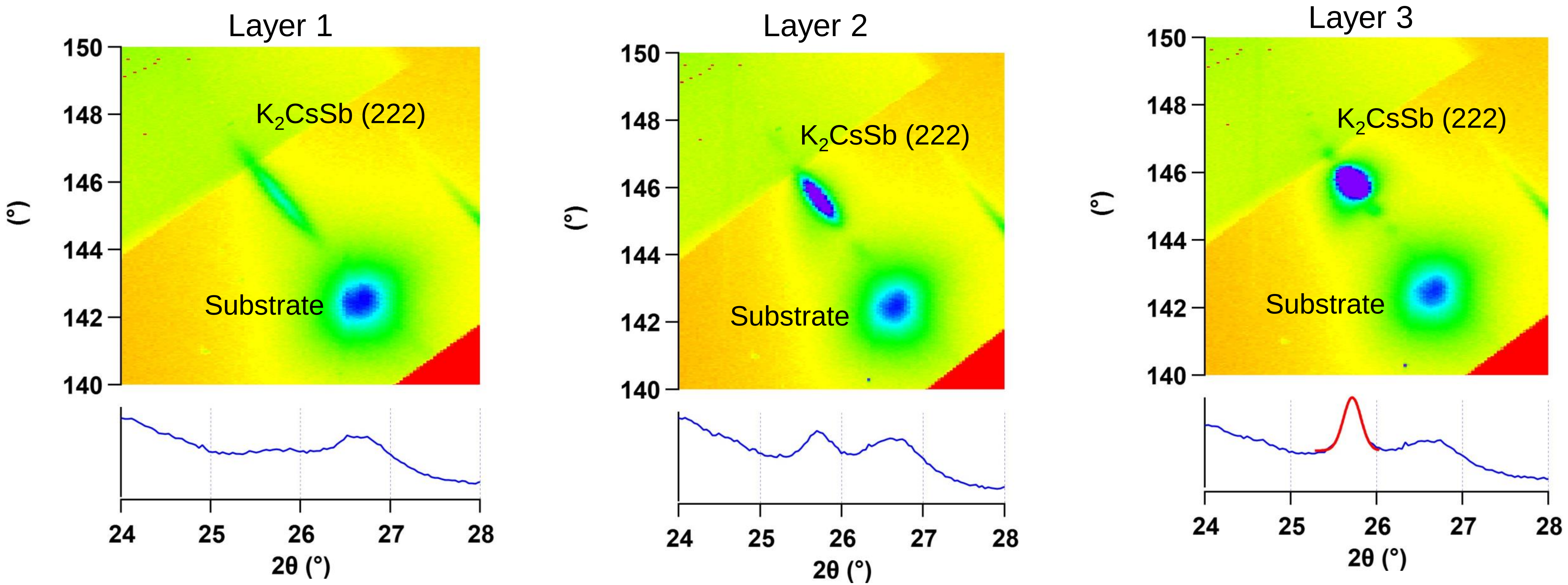




Azimuthal angular dependence observed

Brookhaven National Laboratory

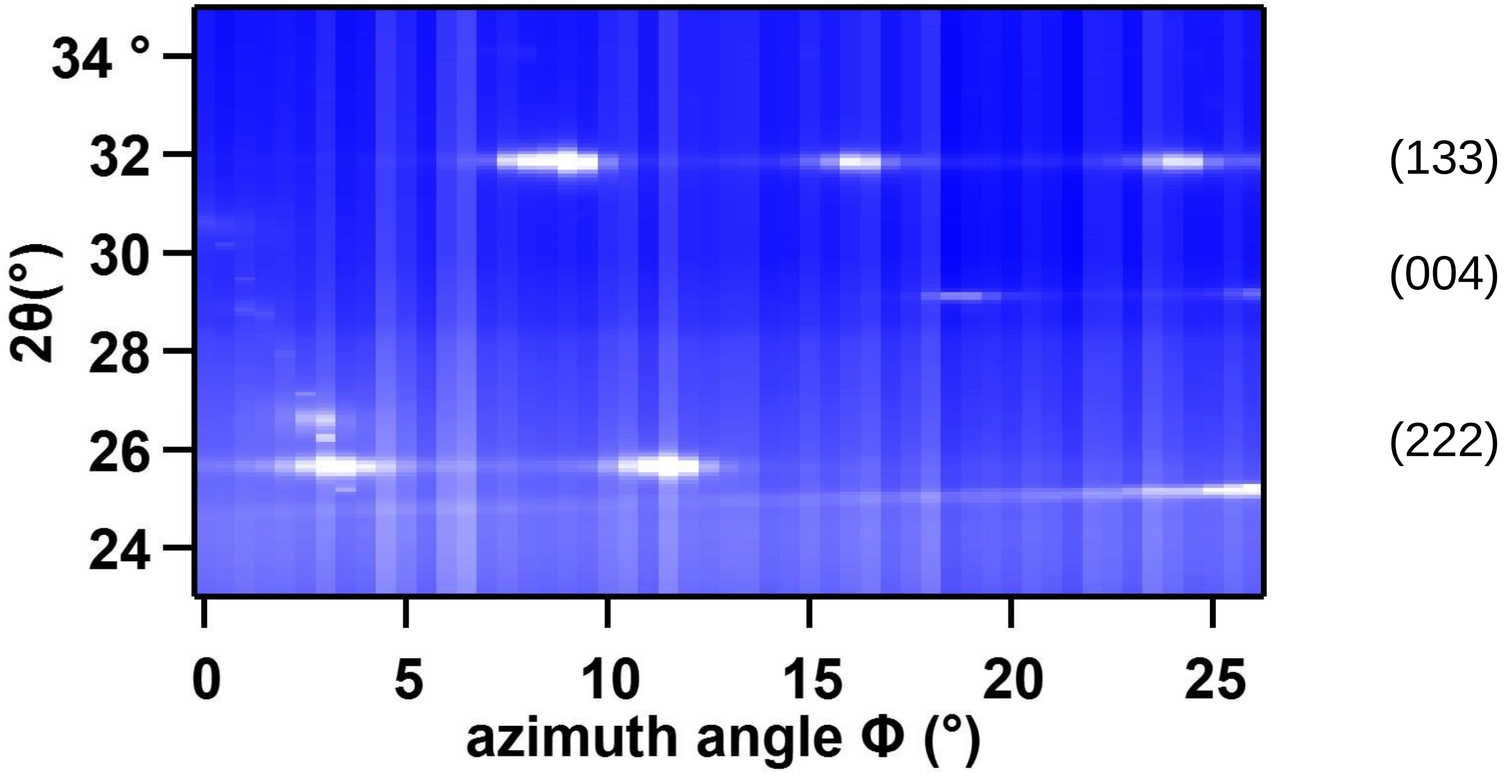
XRD of K_2CsSb/New Substrate (Sample-1): Sb deposited using PLD

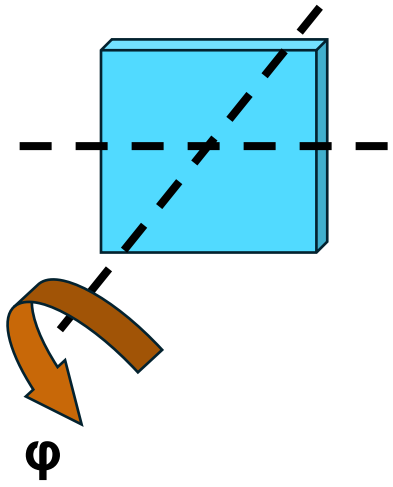


x0 =25.715 ± 0.00224
width =0.14515 ± 0.00415
Grain size: 411.30 nm

- ❑ Diffraction image at Brag condition (222)
- ❑ Elongation structure is from Mosaicity and strain from sample.
- ❑ Grain size of Layer-3 is 411 nm, where film thickness is 20 nm.

Azimuthal angular dependence of XRD from Sample-1





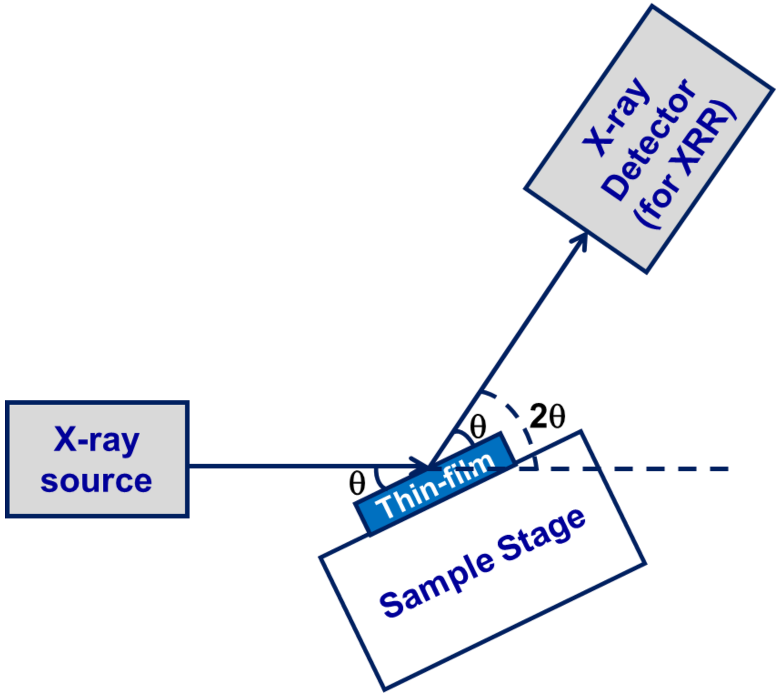
Schematic of azimuthal sample rotation

- ❑ Azimuthal angular dependence observed

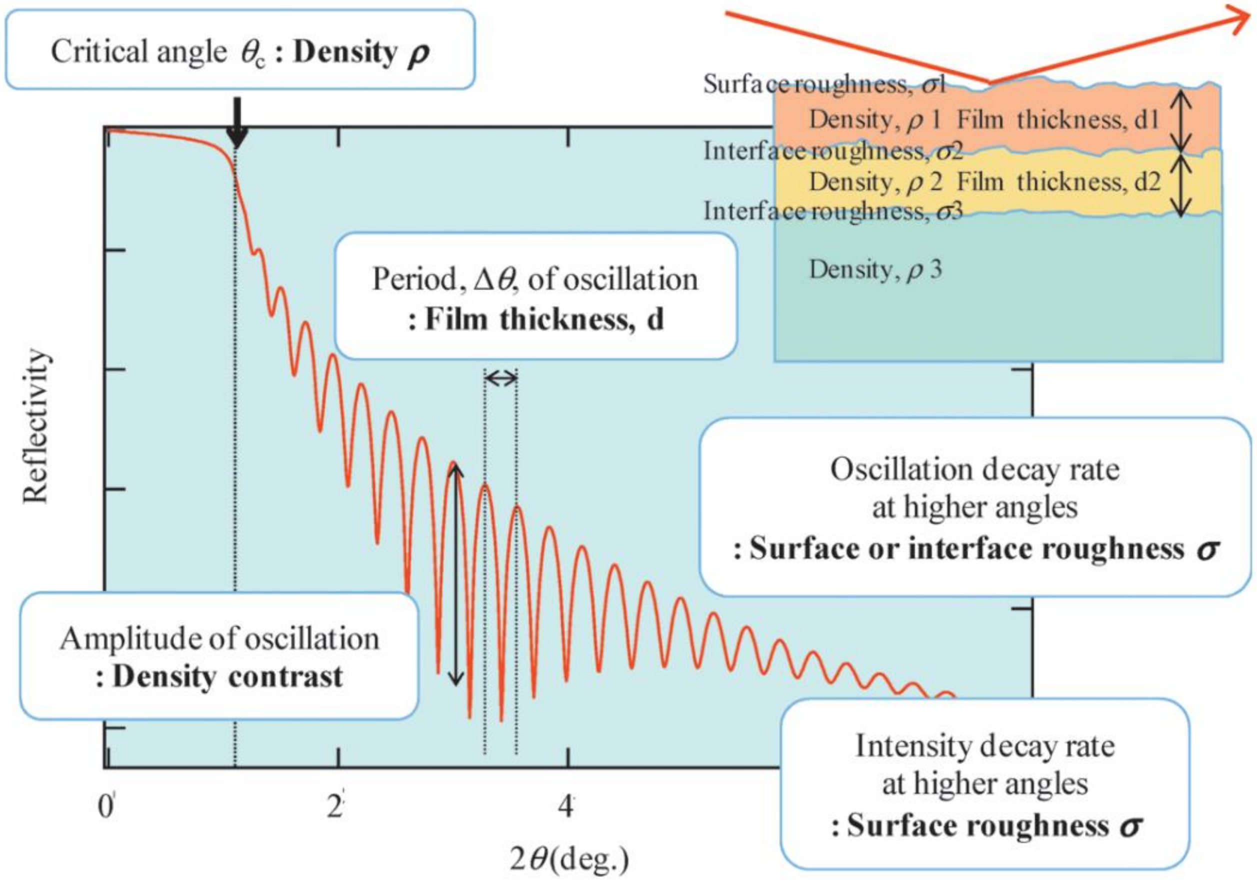
XRR of Photocathode

XRR provides thin-film's

- Film thickness ∝ $1/\Delta\theta$
- Electron density (ρ_e), $\theta_c \propto \sqrt{\rho_e}$
- Roughness, $R_{flat} * \exp(-4kz^2\sigma^2)$

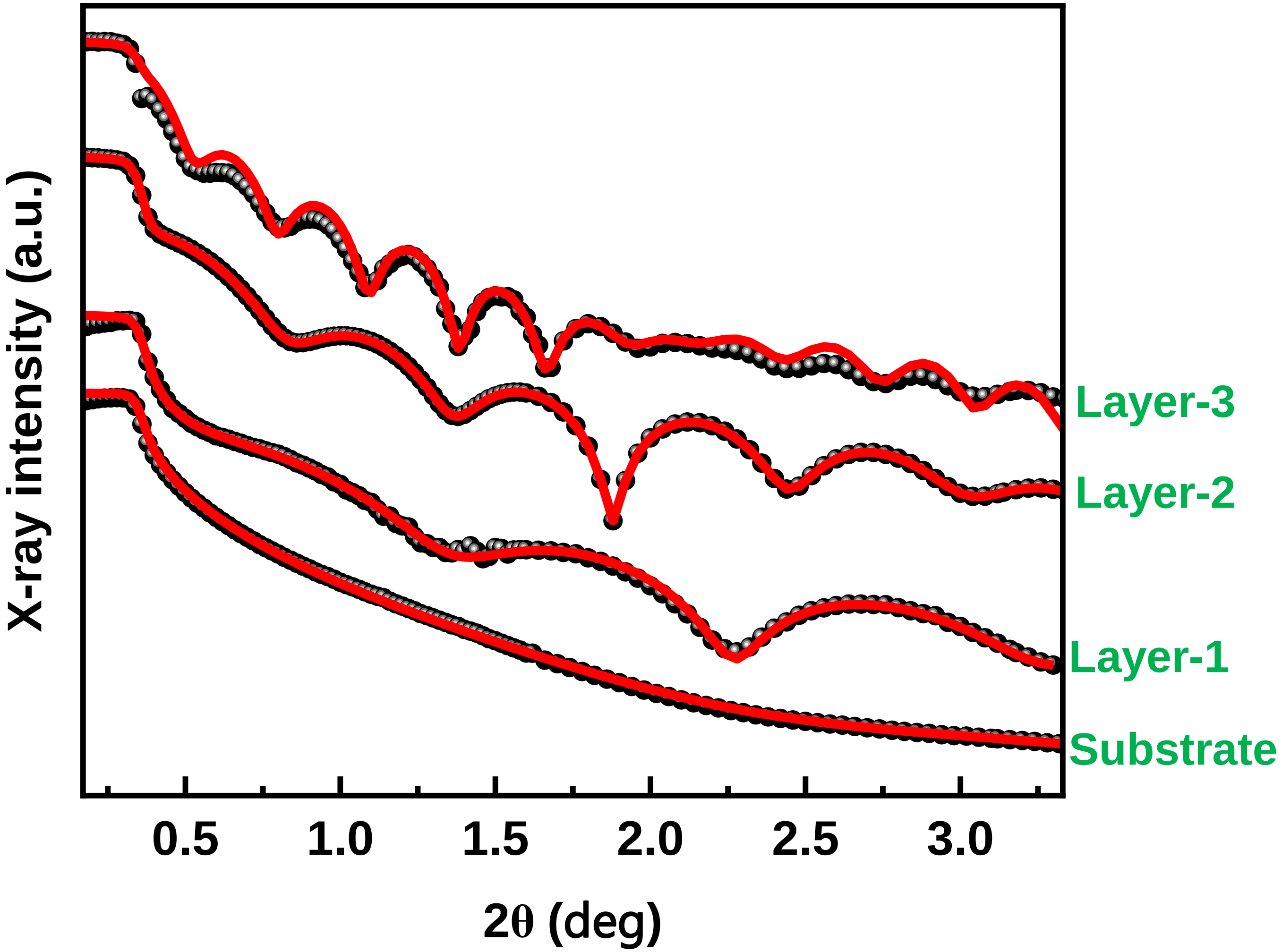


Schematic of XRR



Information provided by XRR [Yasaka, 2010]

XRR of K_2CsSb/New Substrate (Sample-1): Sb deposited using PLD



XRR	Thickness (nm)	Roughness (nm)
Substrate	NA	0.40
Layer 1	6	0.60
Layer 1+2	11	0.65
Layer 1+2+3	20	0.75

XRR of K_2CsSb

- Roughness increased with increasing thickness of photocathode thin film.

Brookhaven National Laboratory

Spectral response of K_2CsSb from Sample 1

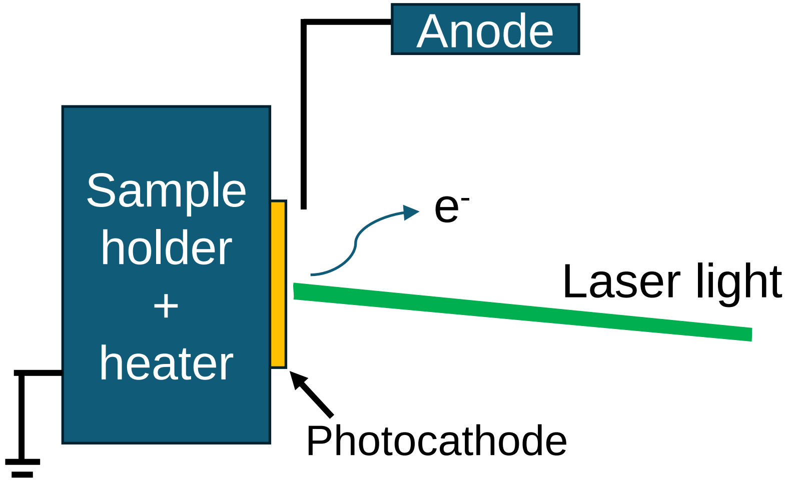
Quantum Efficiency (QE) of Photocathode

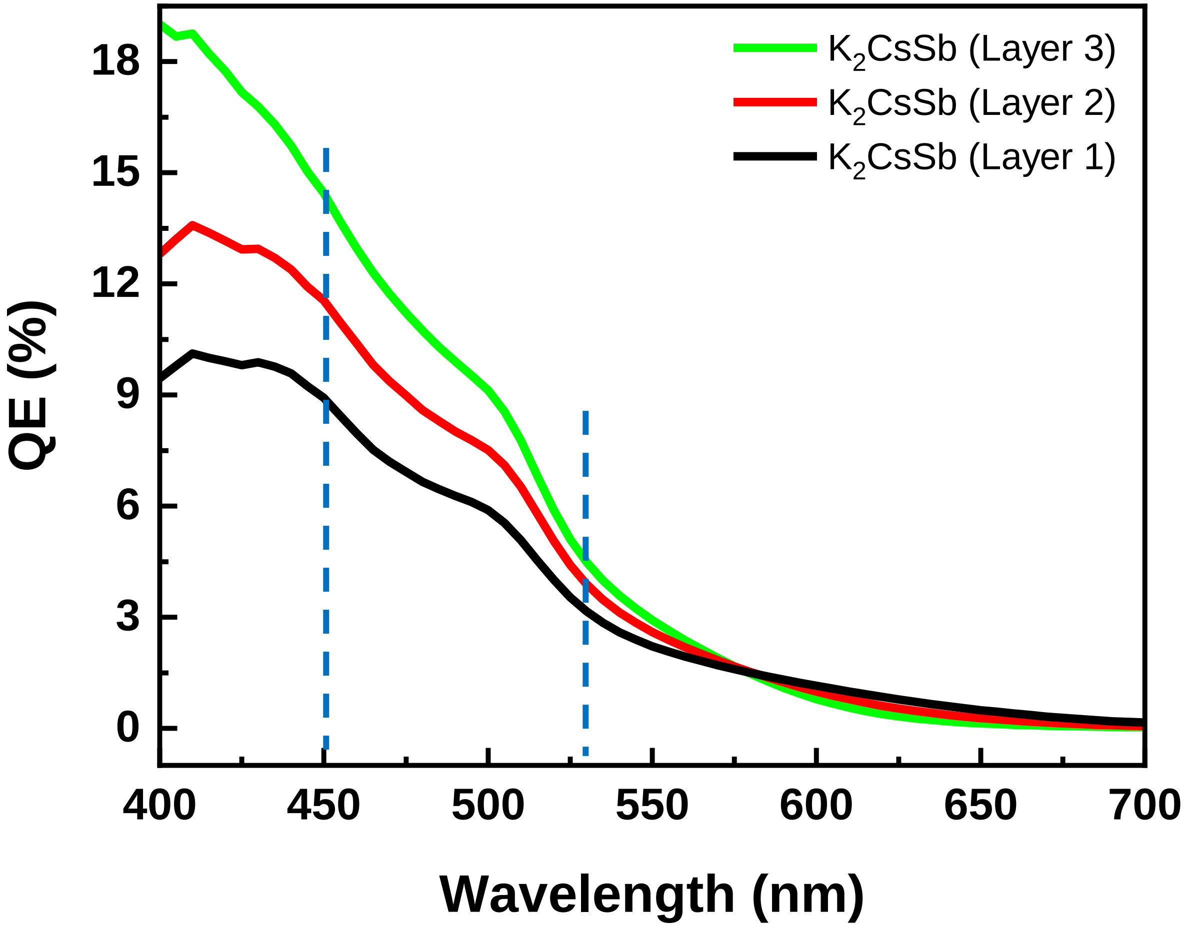
QE is one of the most important parameters, when dealing with photocathodes.

$$QE = \frac{N_{electrons}}{N_{photons}} = \frac{hc}{q_e} \times \frac{I}{\lambda P_{light}}$$

Where I is measured photocurrent from a photocathode, λ is incident wavelength and P_{light} is power of incident light.

Therefore, a maximum QE is achieved by a minimum laser





Wavelength (nm)	QE (%)		
	L1	L2	L3
450 (Blue)	8.9	11.6	14.5
530 (Green)	3.2	3.9	4.5

Brookhaven National Laboratory

PLD assisted epitaxial growth (Sample-1)

- Successful in growing epitaxial thin film photocathode.
- RHEED provides
  - Epitaxial growth of photocathode
  - Photocathode film form is smooth
  - Azimuthal angular dependence observed
- XRR provides
  - Photocathode film thickness, roughness, electron density
  - Film roughness < 0.8 nm
- QE measurements provide QE of photocathode thin film
  - QE for green light 4.5 %.
- XRD provides crystalline structure not only from the surface but also from bulk.

Brookhaven National Laboratory

Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ Experimental Details
- ❑ PLD: epitaxial growth
    - ❑ RHEED, XRD, XRR, GISAXS, XRF results
    - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
    - ❑ RHEED results
    - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

Brookhaven National Laboratory

Sample 1	Sample 2	Sample 3
K_2CsSb	K_2CsSb	K_2CsSb
New Substrate	New Substrate	4H-SiC
❑ Sb deposited using PLD.	❑ Substrate is heat clean from Sample-1. ❑ Sb deposited using Thermal evaporation.	❑ Sb deposited using Thermal evaporation.

For all samples

- ❑ Cs and K were deposited using effusion cells.
- ❑ Layer 1: K_2CsSb on substrate, for structural study
- ❑ Layer 2: Growth of another layer of K_2CsSb on Layer 1
- ❑ Layer 3: Growth of photocathode high enough thickness to achieve high QE.

K_2CsSb (Layer 3)

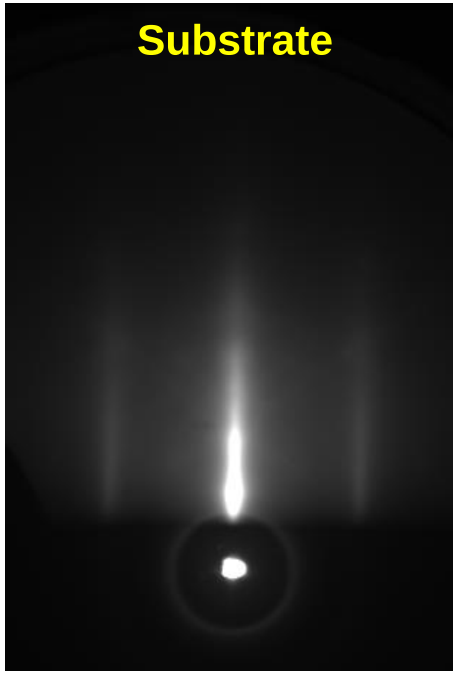
K_2CsSb (Layer 2)

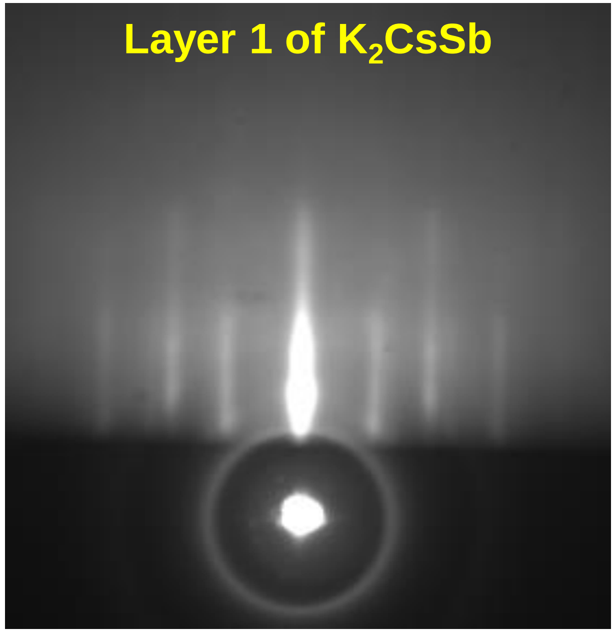
K_2CsSb (Layer 1)

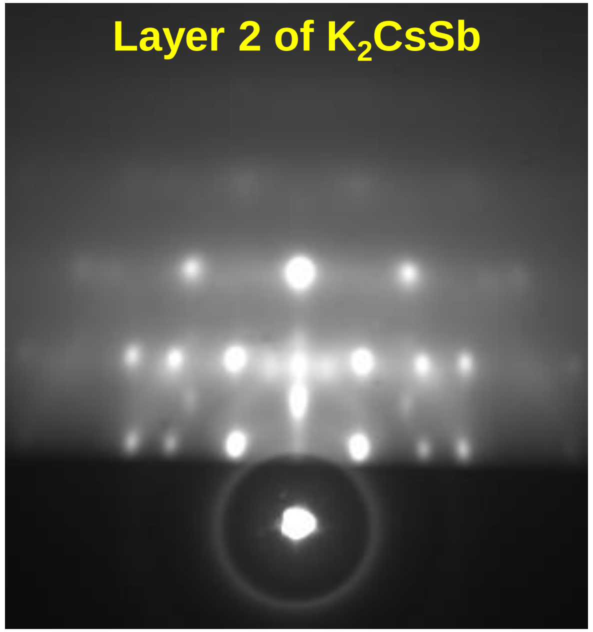
Substrate

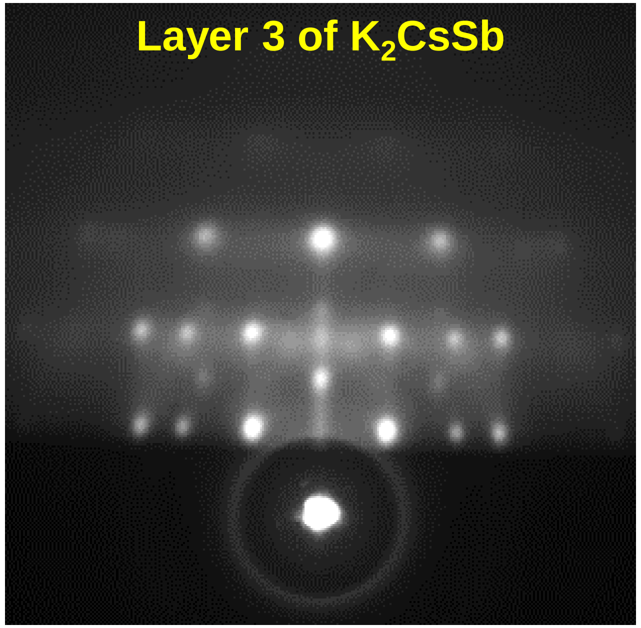
Brookhaven National Laboratory

RHEED of K_2CsSb/New Substrate (Sample-2)









RHEED of K_2CsSb

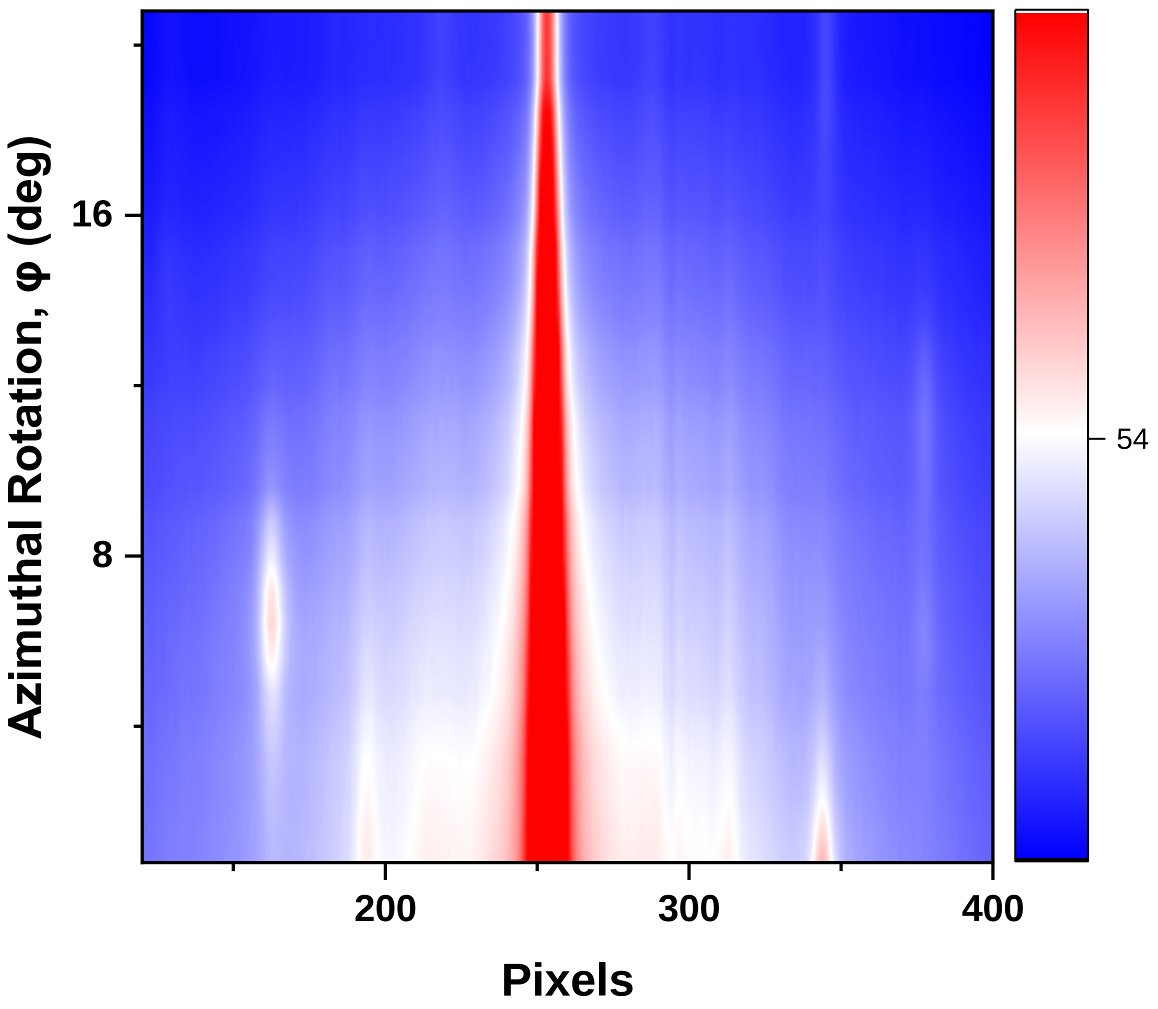
- ❑ RHEED image shows epitaxial growth.
- ❑ Streaks represent smooth surfaces with small domains.
- ❑ Dots represents 3D islands.

K_2CsSb on Substrate			
Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	34.55	4.96	111
2nd	59	2.91	122
3rd	69.25	2.48	222
4th	90.4	1.90	133

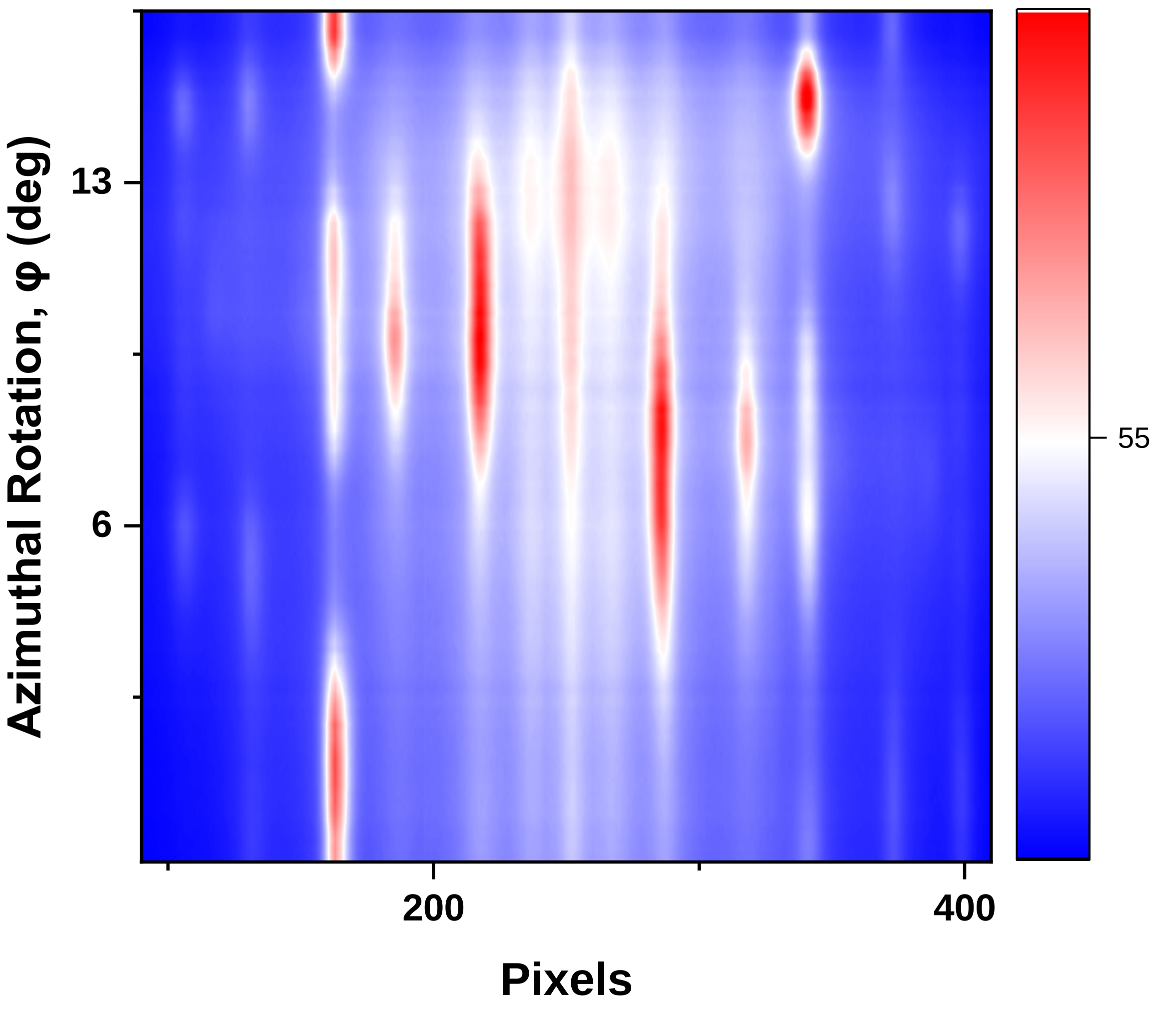
Brookhaven National Laboratory

Azimuthal angular dependence of RHEED from Sample-2

Layer 1 of K_2CsSb



Layer 3 of K_2CsSb



Azimuthal angular dependence observed

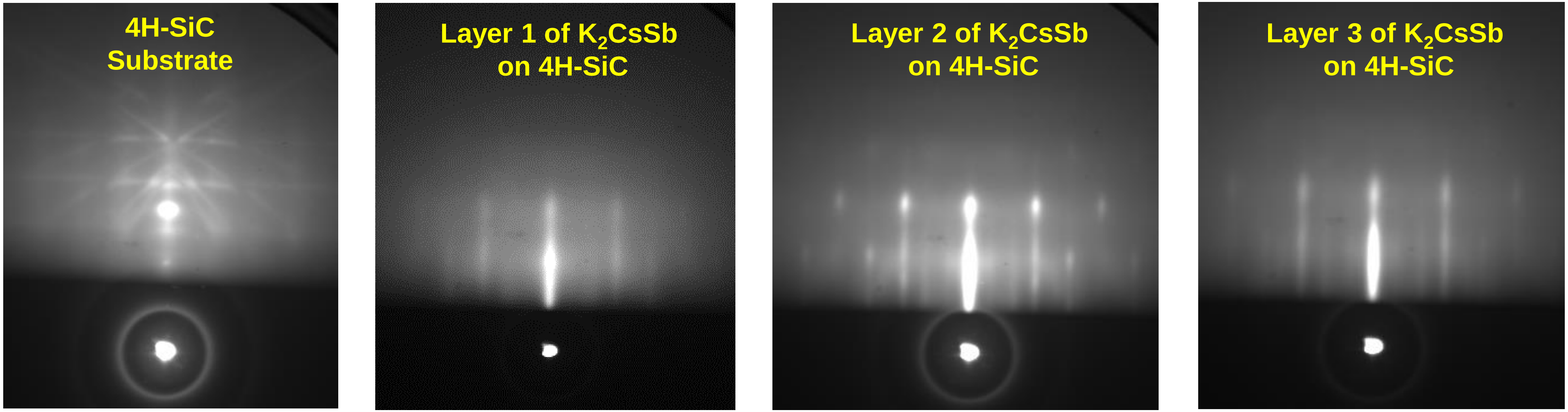
Sample 1	Sample 2	Sample 3
K_2CsSb	K_2CsSb	K_2CsSb
New Substrate	New Substrate	4H-SiC
❑ Sb deposited using PLD.	❑ Substrate is heat clean from Sample-1. ❑ Sb deposited using Thermal evaporation.	❑ Sb deposited using Thermal evaporation.

For all samples

- ❑ Cs and K were deposited using effusion cells.
- ❑ Layer 1: K_2CsSb on substrate, for structural study
- ❑ Layer 2: Growth of another layer of K_2CsSb on Layer 1
- ❑ Layer 3: Growth of photocathode high enough thickness to achieve high QE.

K_2CsSb (Layer 3)
K_2CsSb (Layer 2)
K_2CsSb (Layer 1)
Substrate

RHEED of K_2CsSb/4H-SiC (Sample -3): Sb deposited using thermal evaporation



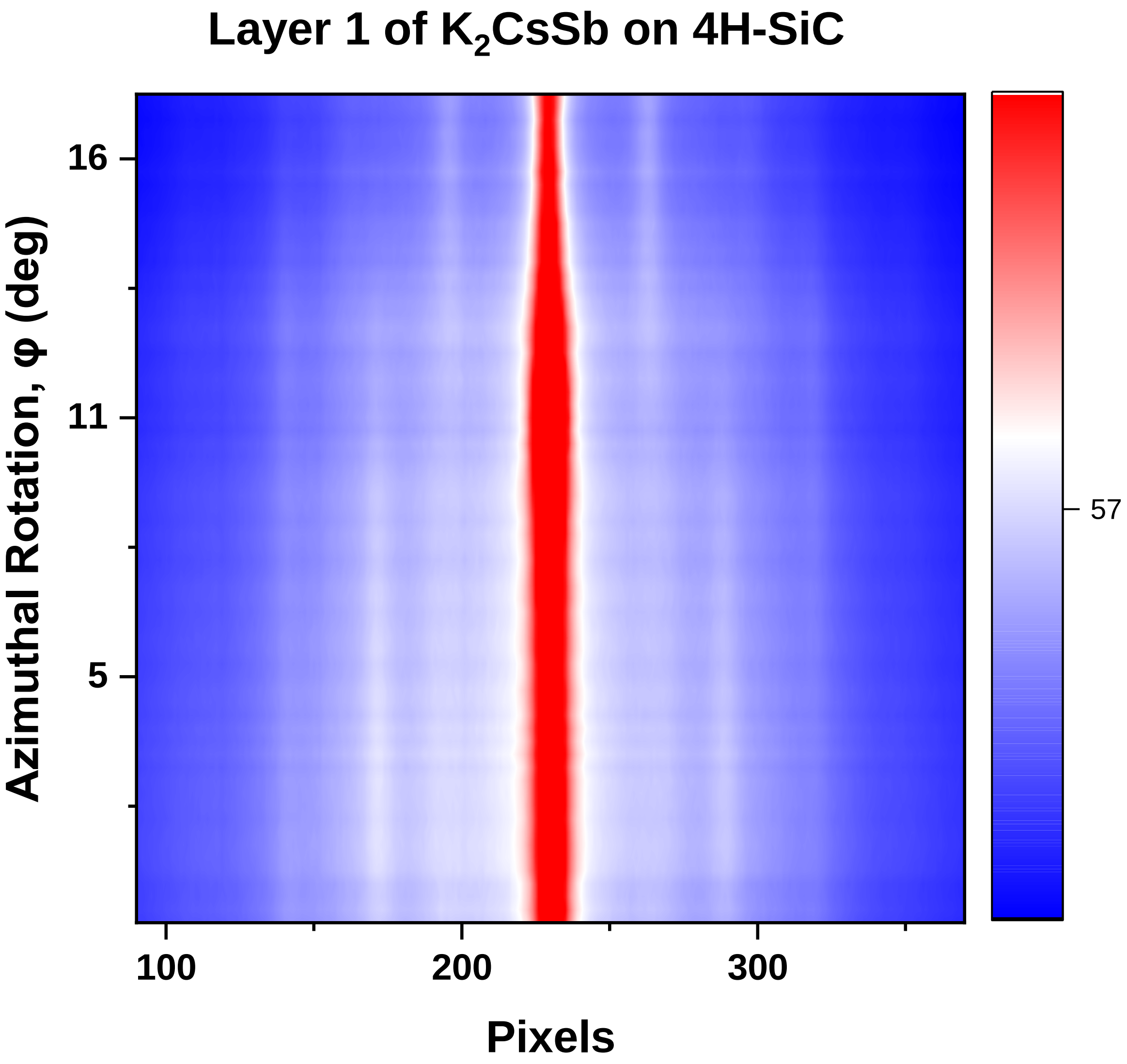
RHEED of K_2CsSb/4H-SiC

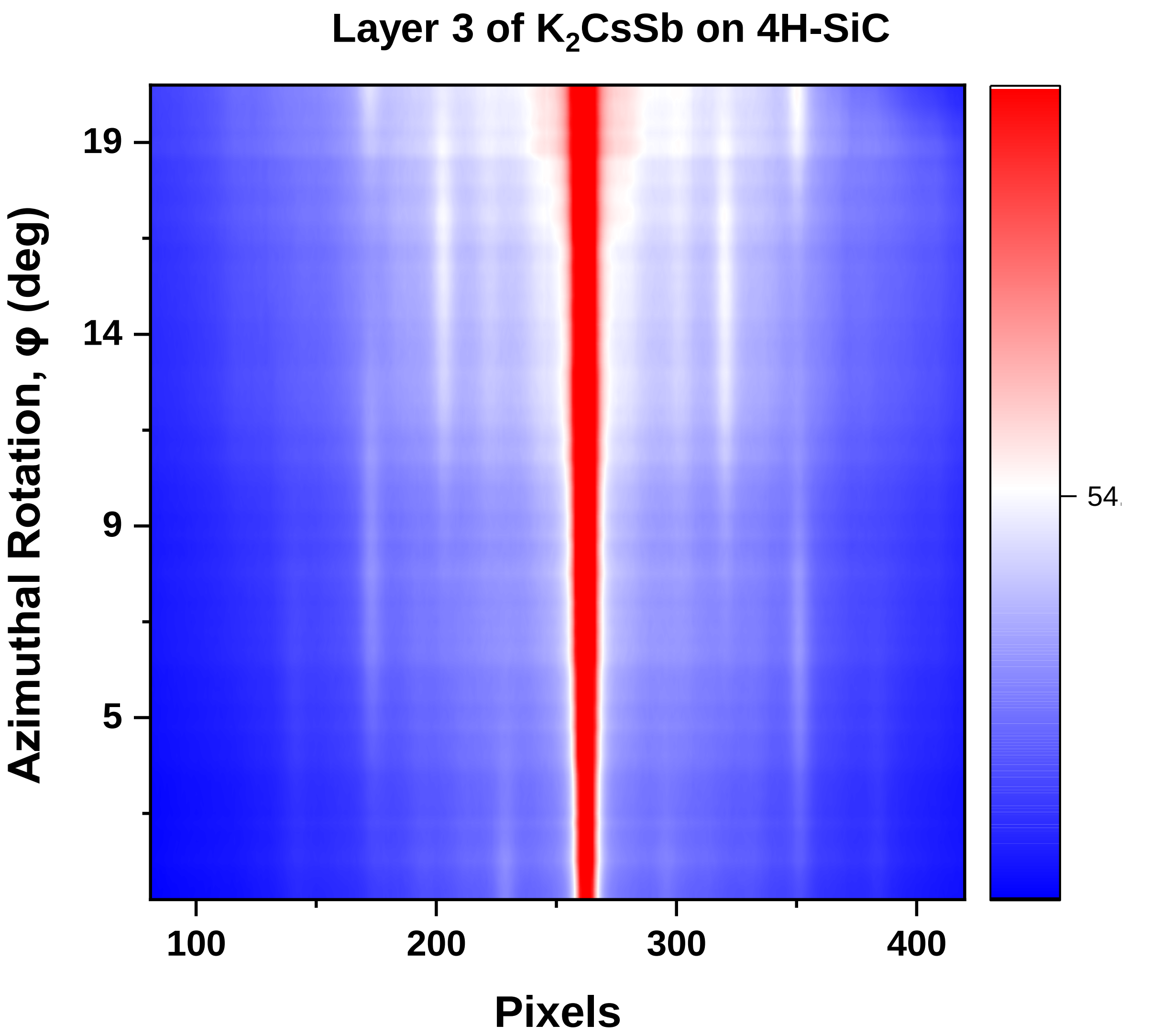
- ❑ RHEED image shows epitaxial growth.
- ❑ Streaks represent smooth surfaces with small domains.

K_2CsSb on 4H-SiC			
Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	38.9	4.41	002
2nd	58.7	2.92	122
3rd	74.75	2.29	004
4th	89.15	1.92	133

Brookhaven National Laboratory

Azimuthal angular dependence of RHEED from Sample-3





Azimuthal angular dependence observed

Compare of RHEED results of K_2CsSb

Conclusion from RHEED

- Similar structure and Sociometry found.
- Thermal evaporation produces rougher surface (need to confirm)

Sample-1: K_2CsSb on New Substrate, Sb deposited using PLD

Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	34.5	4.97	111
2nd	60	2.86	122
3rd	69	2.49	222
4th	91.5	1.87	133

Sample-2: K_2CsSb on New substrate, Sb deposited using Thermal

Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	34.55	4.96	111
2nd	59	2.91	122
3rd	69.25	2.48	222
4th	90.4	1.90	133

Sample-3: K_2CsSb on 4H-SiC, Sb deposited using Thermal evaporation

Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	38.9	4.41	002
2nd	58.7	2.92	122
3rd	74.75	2.29	004
4th	89.15	1.92	133

Brookhaven National Laboratory

Sample 1	Sample 2	Sample 3
K_2CsSb	K_2CsSb	K_2CsSb
New Substrate	New Substrate	4H-SiC
❑ Sb deposited using PLD.	❑ Substrate is heat clean from Sample-1. ❑ Sb deposited using Thermal evaporation.	❑ Sb deposited using Thermal evaporation.

For all samples

- ❑ Cs and K were deposited using effusion cells.
- ❑ Layer 1: K_2CsSb on substrate, for structural study
- ❑ Layer 2: Growth of another layer of K_2CsSb on Layer 1
- ❑ Layer 3: Growth of photocathode high enough thickness to achieve high QE.

K_2CsSb (Layer 3)

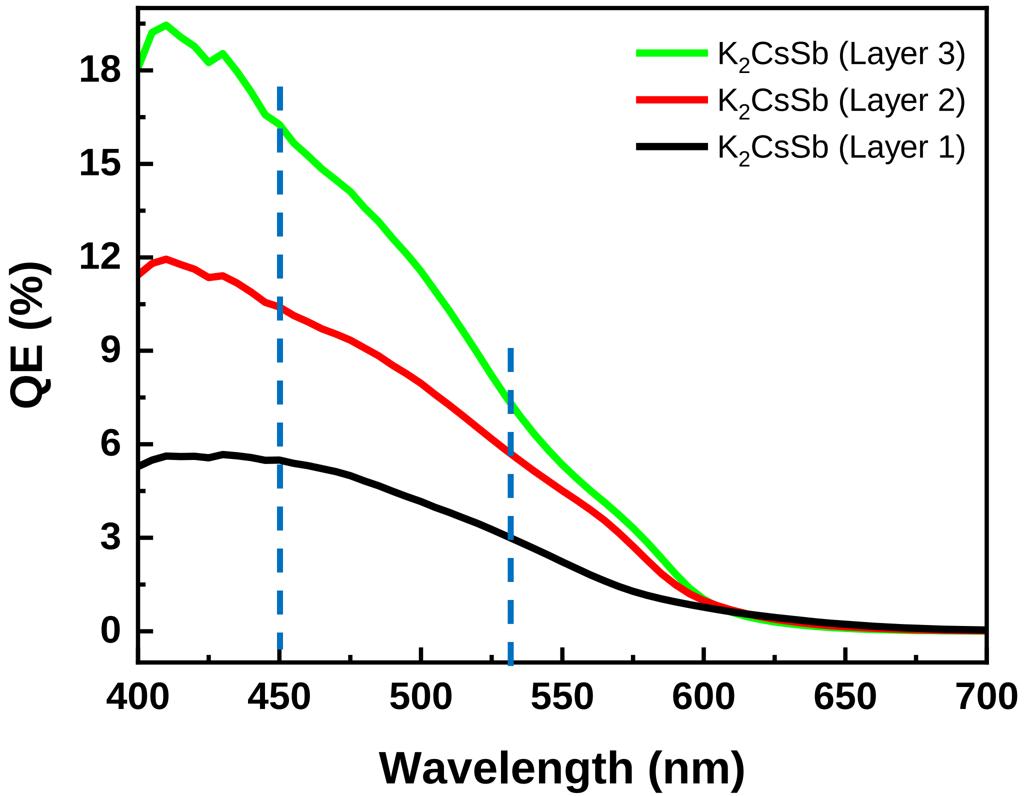
K_2CsSb (Layer 2)

K_2CsSb (Layer 1)

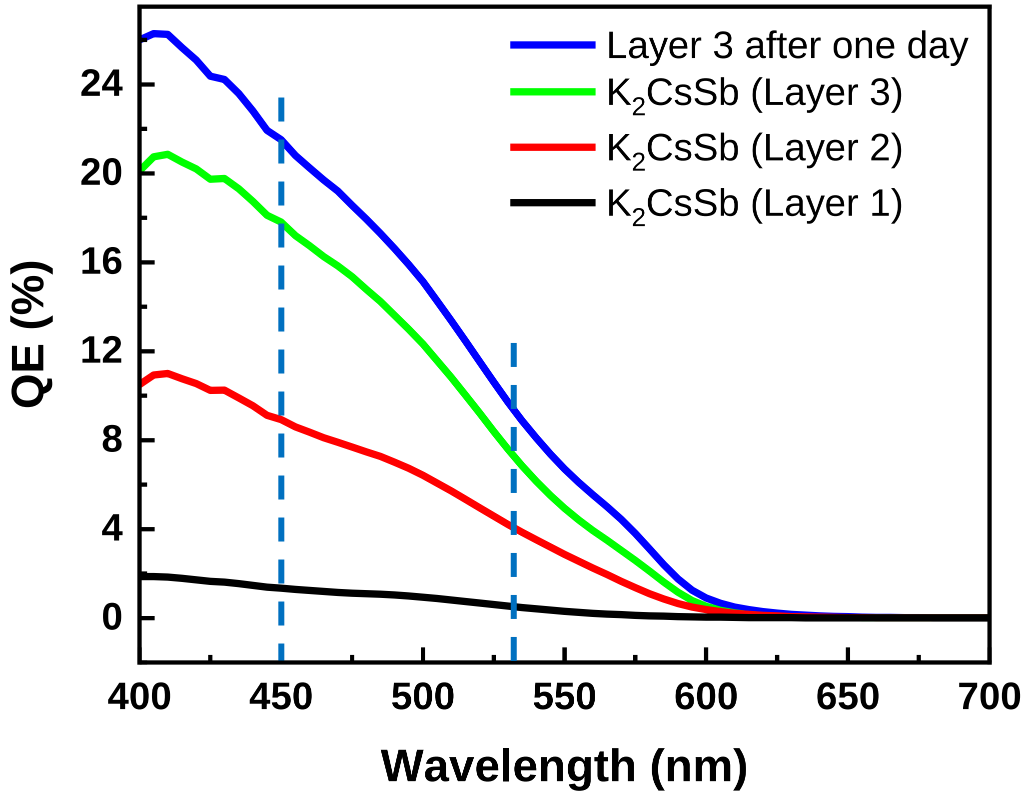
Substrate

Brookhaven National Laboratory

Spectral response from Sample 2



Spectral response from Sample 3



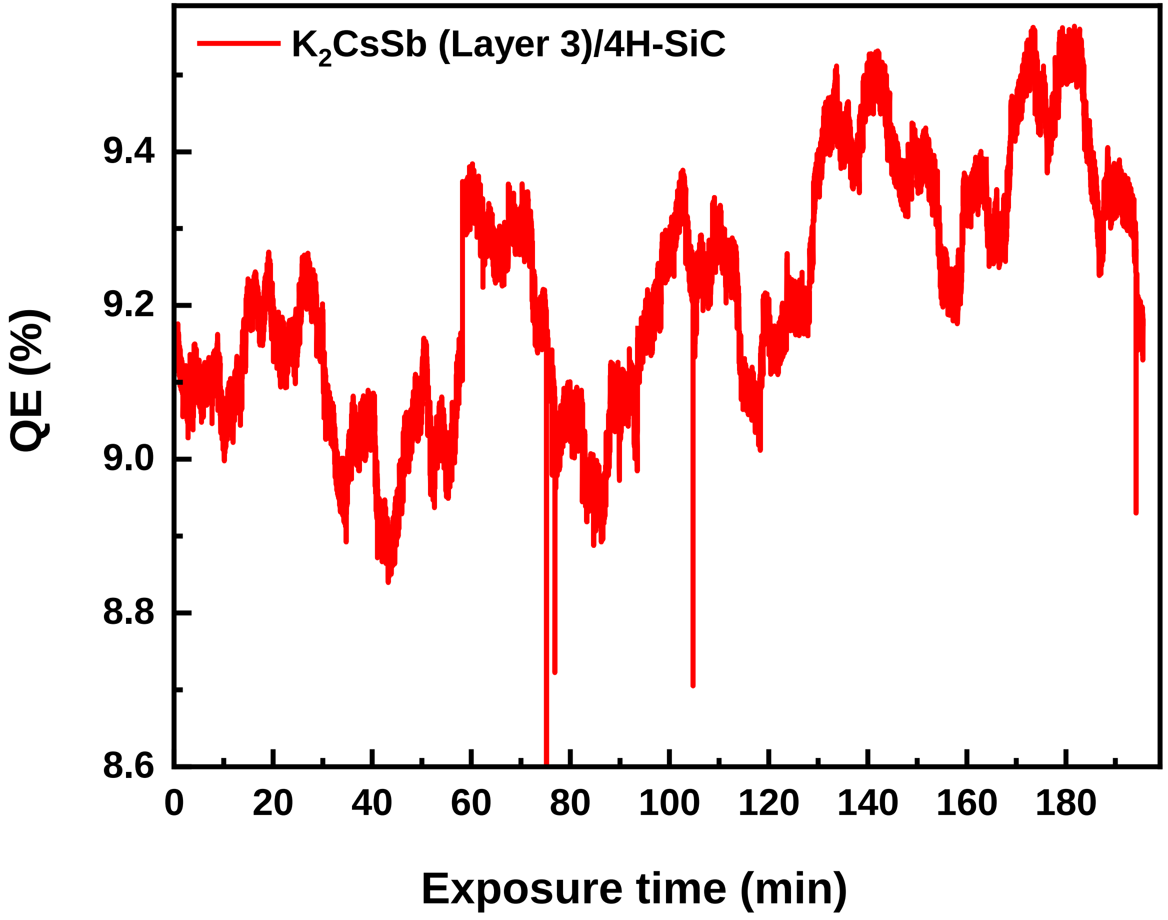
Wavelength (nm)	Sample-2 QE (%)		
	L1	L2	L3
450 (Blue)	5.5	10.4	16.3
530 (Green)	3.1	5.8	7.5

Wavelength (nm)		Sample-3 QE (%)			
		L1	L2	L3	L3 after 1 day
Green laser		0.5	4.6	9.2	--
LDLS source	450	1.3	8.9	17.8	21.5
	530	0.5	4.2	7.6	9.7

Brookhaven National Laboratory

Lifetime of K_2CsSb/4H-SiC using Green laser

- Measured QE after 3-4 days
- Current draw about 25 uA
- No decay in QE
- Fluctuations from laser power



Brookhaven National Laboratory

Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ Experimental Details
- ❑ PLD: epitaxial growth
  - ❑ RHEED, XRD, XRR, GISAXS, XRF results
  - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
  - ❑ RHEED results
  - ❑ Spectral response
- ❑ **Conclusions**
- ❑ Acknowledgement

Brookhaven National Laboratory

Conclusion

- Successful in growing epitaxial thin film of photocathode.
- RHEED provides
  - Epitaxial growth of photocathode
  - Photocathode films formed are smooth surface
  - Azimuthal angular dependence observed
  - PLD and thermal grow similar structure
- XRR provides
  - Photocathode film thickness, roughness, electron density
  - Film roughness < 0.8 nm
- QE measurements provide QE of photocathode thin film
  - QE got for green laser > 9%.
- XRD provides crystalline structure not only from the surface but also from bulk
  - Azimuthal angular dependence observed

Outline

- ❑ Introduction: Photocathode and epitaxial growth
- ❑ Experimental Details
- ❑ PLD: epitaxial growth
  - ❑ RHEED, XRD, XRR, GISAXS, XRF results
  - ❑ Spectral response
- ❑ Thermal: Epitaxial growth
  - ❑ RHEED results
  - ❑ Spectral response
- ❑ Conclusions
- ❑ Acknowledgement

Acknowledgements

Work supported by Brookhaven Science Associates, LLC under Contract 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.

Brookhaven National Laboratory

Acknowledgment

Brookhaven National Laboratory

Mengjia Gaowei (Supervisor), Pallavi Saha, Kenneth Evans-Lutterodt, Raul Acevedo-Esteves, Jean Jordan-Sweet, John Walsh, Rudy Begay, Jyoti Biswas, Luca Cultrera

SLAC National Accelerator Laboratory, Los Alamos National Laboratory

John Smedley

Cornell University

Chad Pennington, Elena Maria Echeverria Mora, Jared Maxson

Arizona State University

Priyadarshini Bhattacharyya, Siddharth Karkare

Leiden Institute of Physics

Molen S.J. Van der, Guido Stam

University of Vienna

Thomas Juffmann

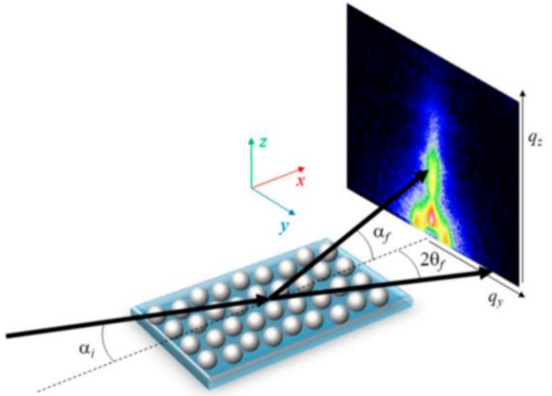
IBM T.J. Watson Research Center

Rudolf Tromp

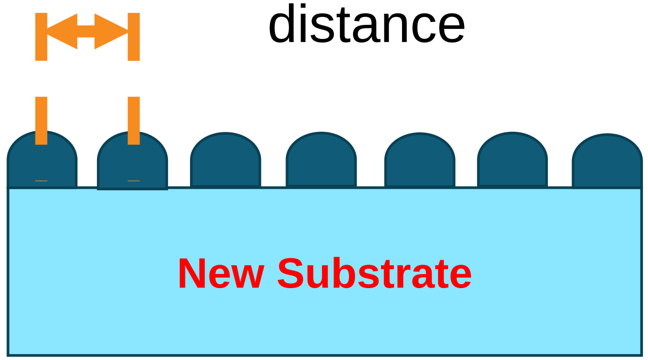
Thank You

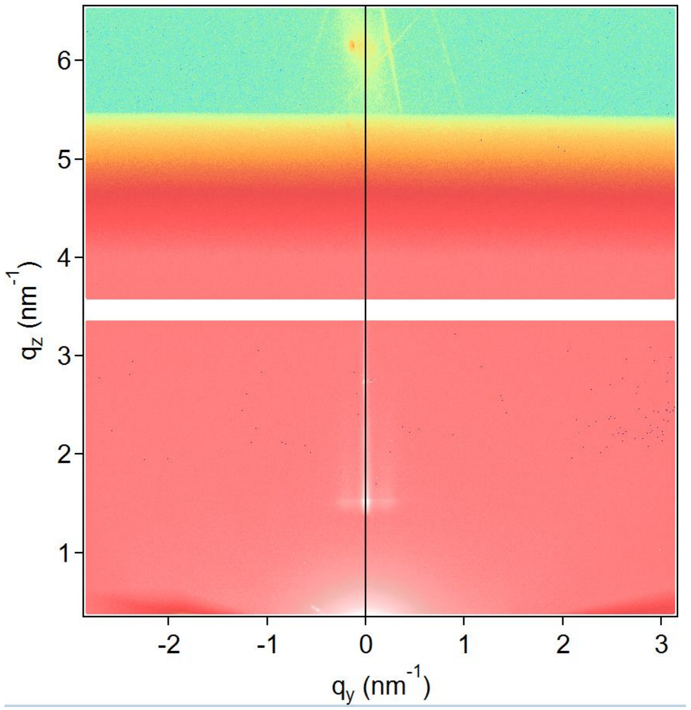
GISAXS: Photocathode on single crystal

- Morphology and distribution of either islands on a substrate or buried particles.
- Structural details of thin film like correlations, Shape, Size of density inhomogeneities at surfaces or at buried interfaces.
- Depth sensitivity from surface to buried interfaces up to a few hundreds of nanometers can be probed by varying the angle of incidence.

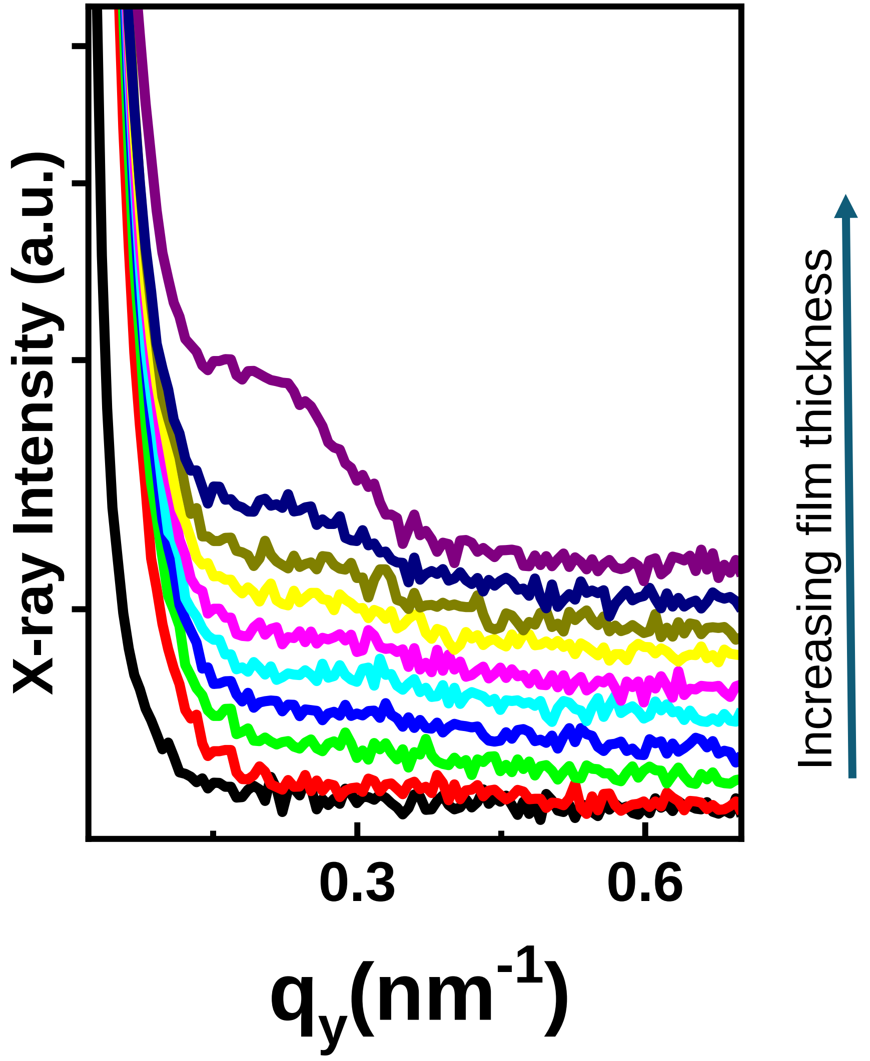


L , Average particle distance





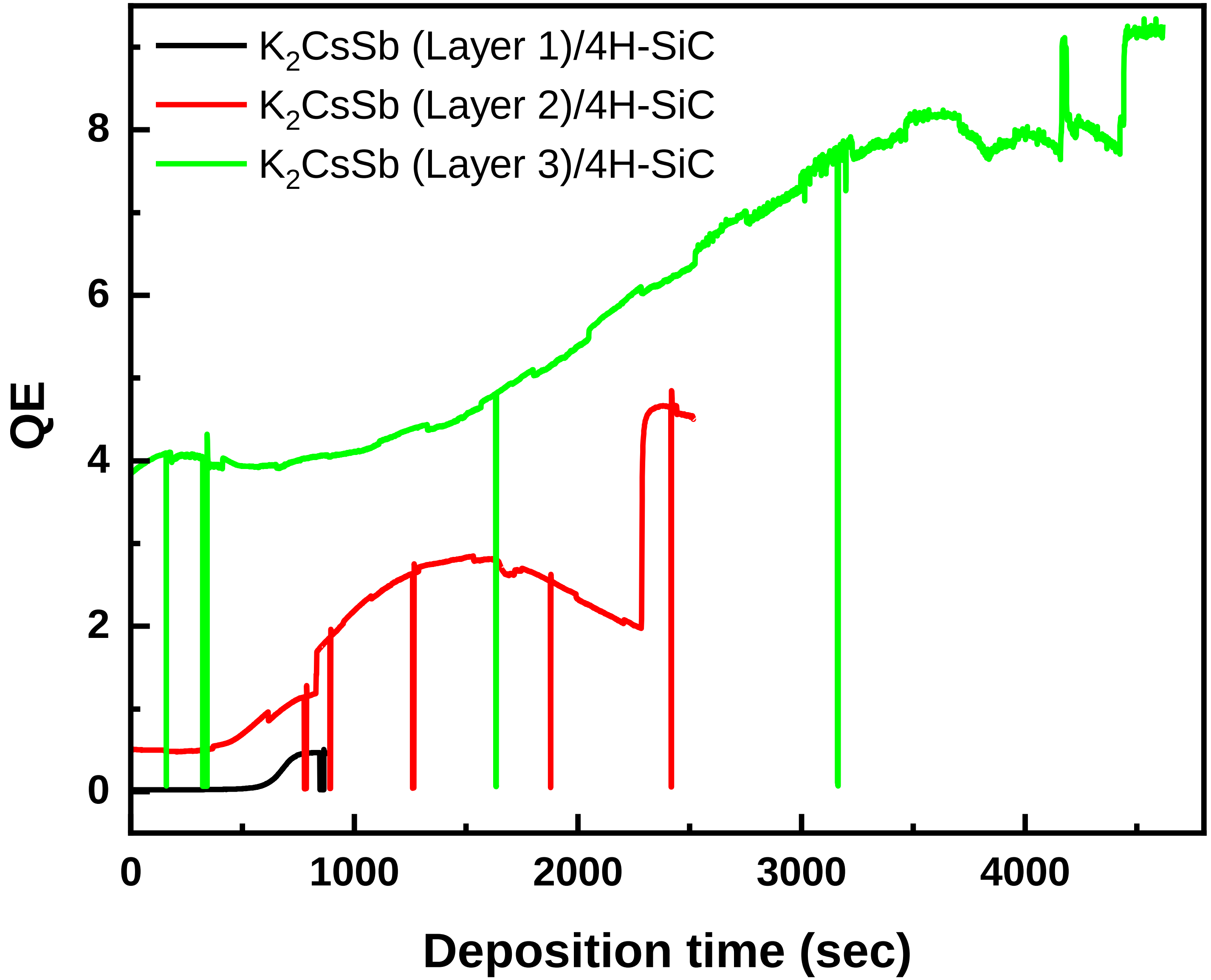
GISAXS Image



GISAXS of K_2CsSb

- Layer 1: average particle distance 35 nm.

Brookhaven National Laboratory



Brookhaven National Laboratory