Pulsed laser deposition assisted epitaxial growth of cesium telluride photocathode for high brightness electron source

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Outline

- Introduction: Photocathode and epitaxial growth
- Experimental Details
- PLD: epitaxial growth of Cs_2Te
  - RHEED, XRD, XRR, XRF results
  - Spectral response
- Conclusions and Future Plan
- Other Epitaxially grown photocathode Results
- Acknowledgement

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Introduction

- In modern technologies, electron sources have wide range of applications: microscopes, medical diagnostic devices, radio transmitters, etc. It also has wide applications: particle accelerators, photon detectors, X-ray free electron laser (XFEL).

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.

Brightness

- High brightness, characterized by high beam current with low emittance, is essential for these applications. The normalized beam brightness is defined as.

$$B_n = \frac{2I}{\pi^2 \varepsilon_{n,x} \varepsilon_{n,y}}$$; where I is electron beam current, $\varepsilon_{n,x}$ and $\varepsilon_{n,y}$ are normalized transverse emittance.

- High current and brightness require photocathode having high quantum efficiency (QE) at convenient laser wavelengths, low emittance, long lifetime, fast response time, etc.
- 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 cesium telluride (Cs_2Te), bi-alkali antimonide (K_2CsSb), Cesium antimonide (Cs-Sb) or gallium arsenide (GaAs).

- Compared to the traditional sequential deposition, the co-evaporation method is reported to yield better surface roughness, film crystallinity, and high quantum efficiency for photocathode materials [Gaowei *et al* (2019), Ding *et al* (2017)]
- QE of 10% or more at 266 nm is observed with sequential deposition of Cs and Te [Kong *et al* (1995)]
- Co-deposition of Cs-Te provide a QE of 19% at 266 nm with a smooth cathode surface of ~2 nm roughness at a film thickness of over 100 nm [Gaowei *et al* (2019)]

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Epitaxial growth of Photocathode

- Epitaxial growth with lattice matching substrate has become a popular approach to produce ultrasmooth and single-crystal photocathodes for high-brightness electron source applications [Parzyck et al (2022), Guido et al (2024)]
- 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.
- The emitted beam brightness can be limited by the surface and bulk disorder of the polycrystalline photocathode material.
- 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.
- Also, growth of photocathode with low emittance.

Introduction

- Epitaxial growth of Cs_2Te, deposited on different substrates: 4H-SiC, Graphene (Gr)/4H-SiC, $Gr/SiO_2/Si$ [Mondal *et al* (under submission)]
- Epitaxial growth: Pulsed laser deposition assisted growth.

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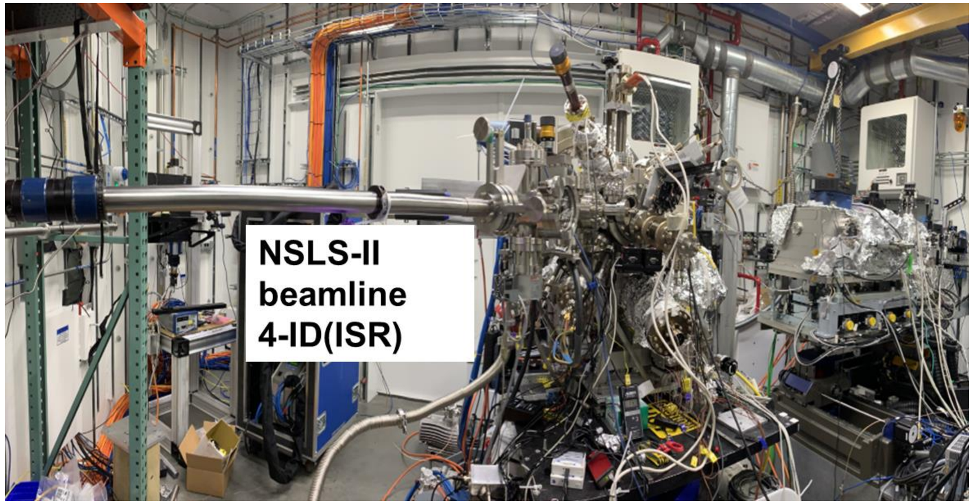
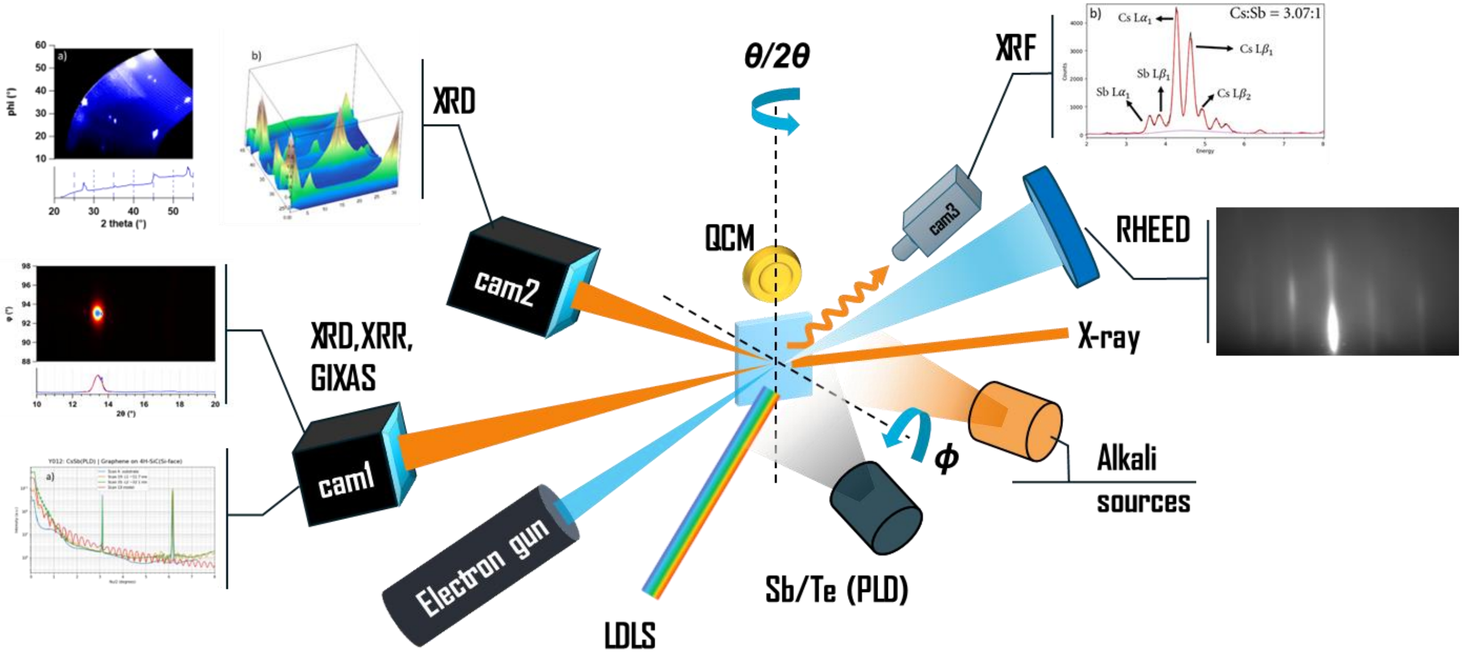
The BNL UHV photocathode growth system at beamline 4-ID, Integrated In situ and Resonant Hard X-ray Studies (ISR), NSLS-II for in situ and real time x-ray characterization. schematic of X-ray techniques (XRR, XRD, GISAXS, XRF) used to characterize the photocathode properties.





Evaporators:

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

- ❑ Cs deposited using effusion cell.
- ❑ Te deposited with using PLD

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

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Epitaxial Growth of Cs_2Te

PLD assisted epitaxial growth

at 4ID, NSLS-II, BNL

Sample 1	Sample 2	Sample 3
Cs_2Te	Cs_2Te	Cs_2Te
4H-SiC	Gr/4H-SiC	Gr/SiO_2/Si

- Gr is epitaxially grown on 4H-SiC.
- Gr transferred on SiO_2 (300nm)/Si.

For all samples

- Te was deposited using PLD and Cs was deposited using effusion cells.
- Layer 1: Cs_2Te on substrate, for structural study with low enough film thickness to confirm epitaxial growth.
- Layer 2: Growth of another layer of Cs_2Te to check the continuity of epitaxial growth
- Layer 3: Growth of photocathode with high enough thickness to achieve high QE.

Cs_2Te (Layer 3)
Cs_2Te (Layer 2)
Cs_2Te (Layer 1)
Substrate

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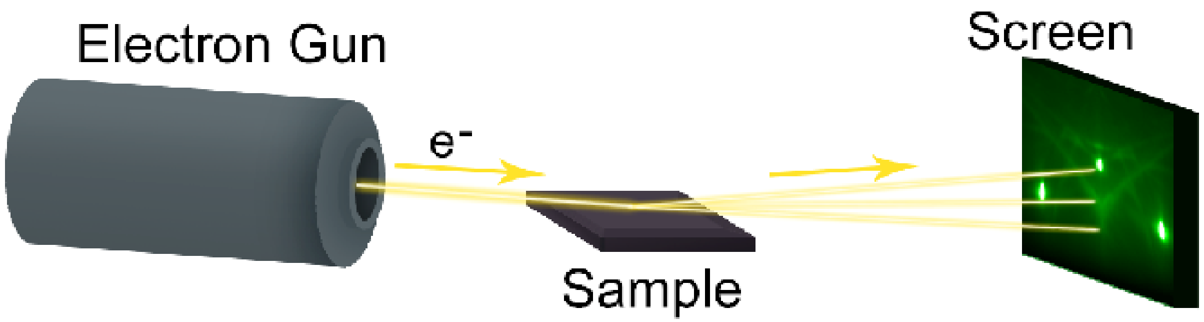
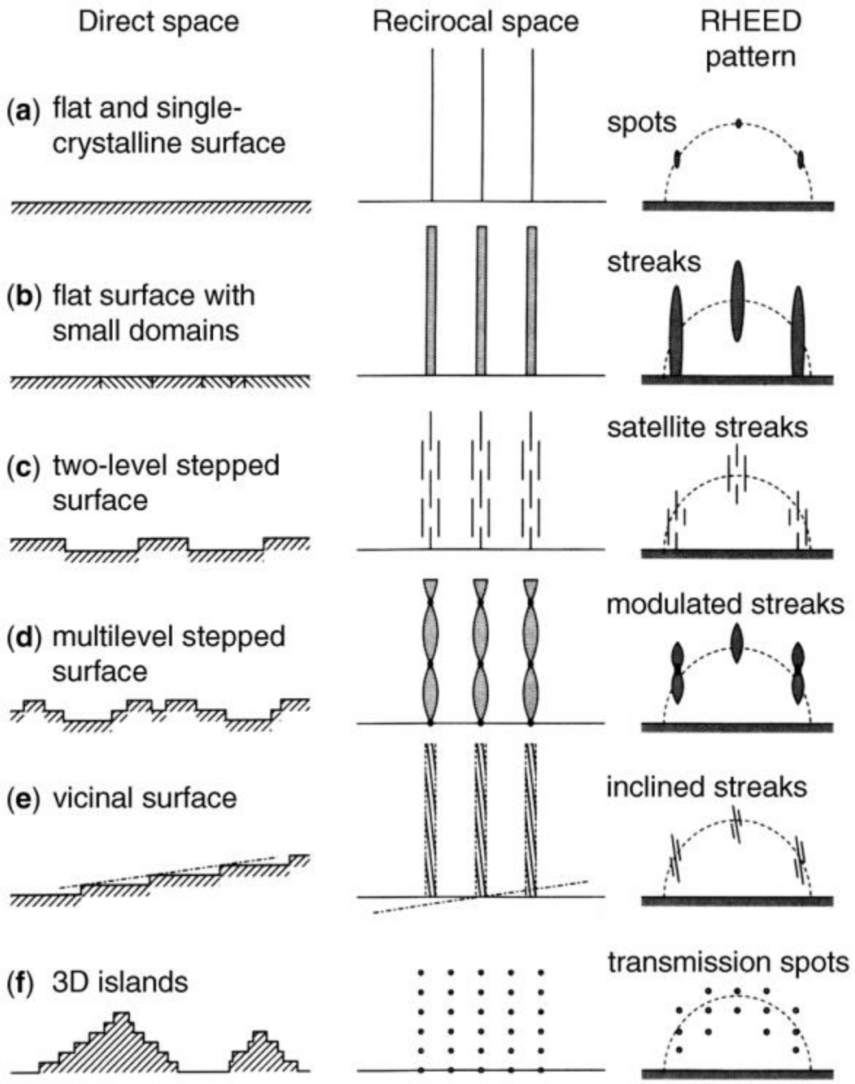
RHEED of Photocathode

- RHEED provides -surface symmetry, real space lattice spacing, crystalline degree of perfection
- Photocathodic thin film, Cs_2Te grown on 4H-SiC, Gr/4H-SiC, $Gr/SiO_2/Si$.
- 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]

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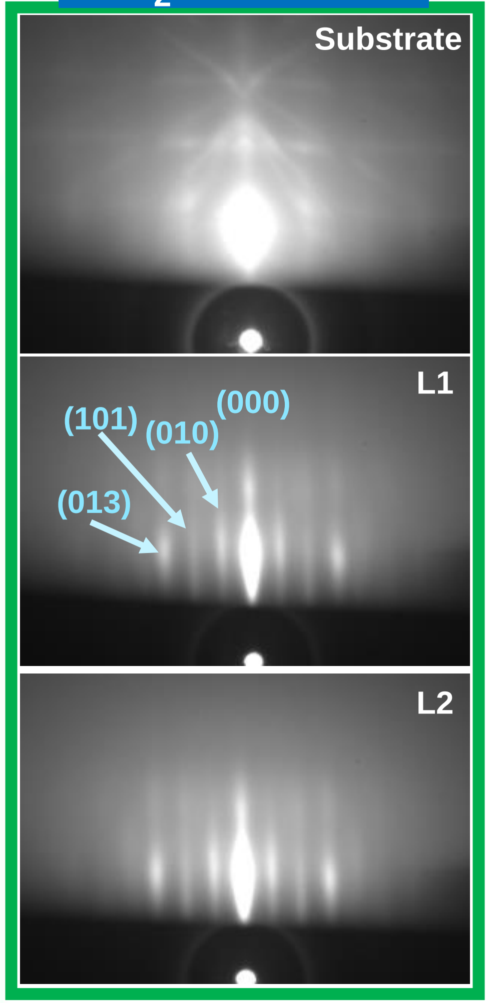
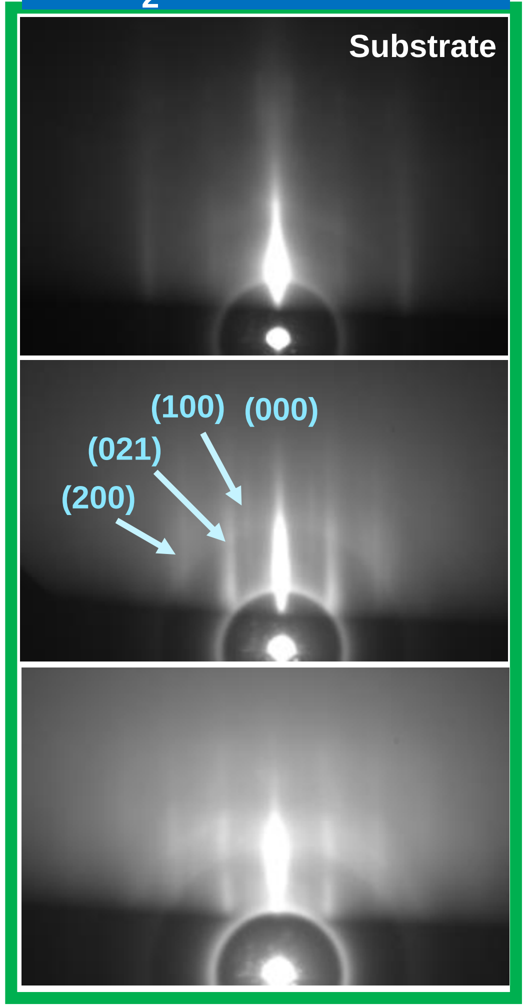
RHEED of Cs_2Te grown on different substrate

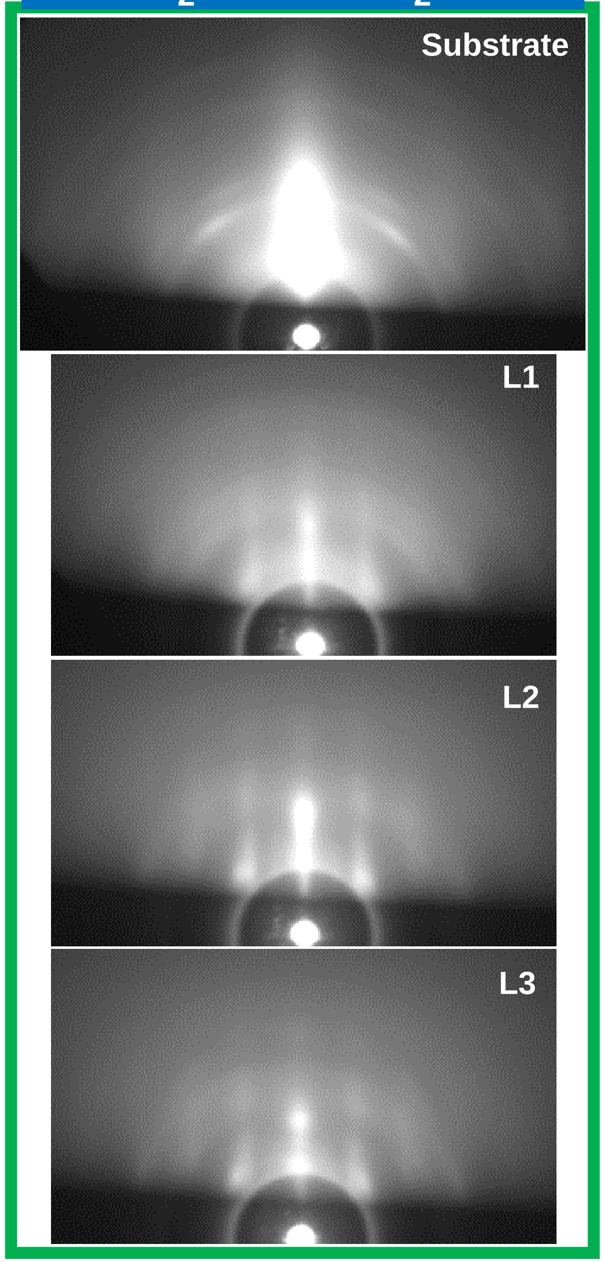
Cs_2Te/4H-SiC



Cs_2Te/Gr/4H-SiC



Cs_2Te/Gr/SiO_2/Si



- The streaky RHEED indicates flat, smooth surfaces with small domains, characteristic of highly crystalline films on 4H-SiC and Gr/4H-SiC substrates.
- Both streaky and ring patterns, suggesting an initial formation of textured or polycrystalline Cs_2Te film on transfer Gr on SiO_2/Si substrate.

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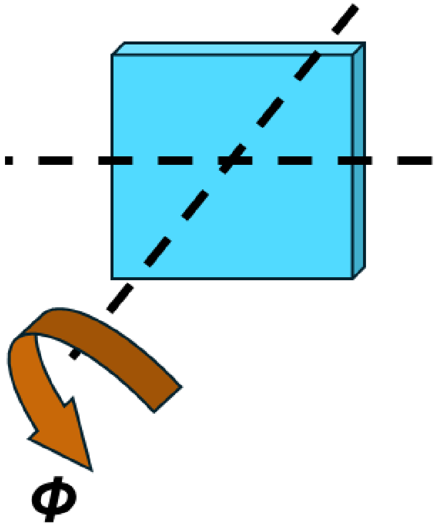
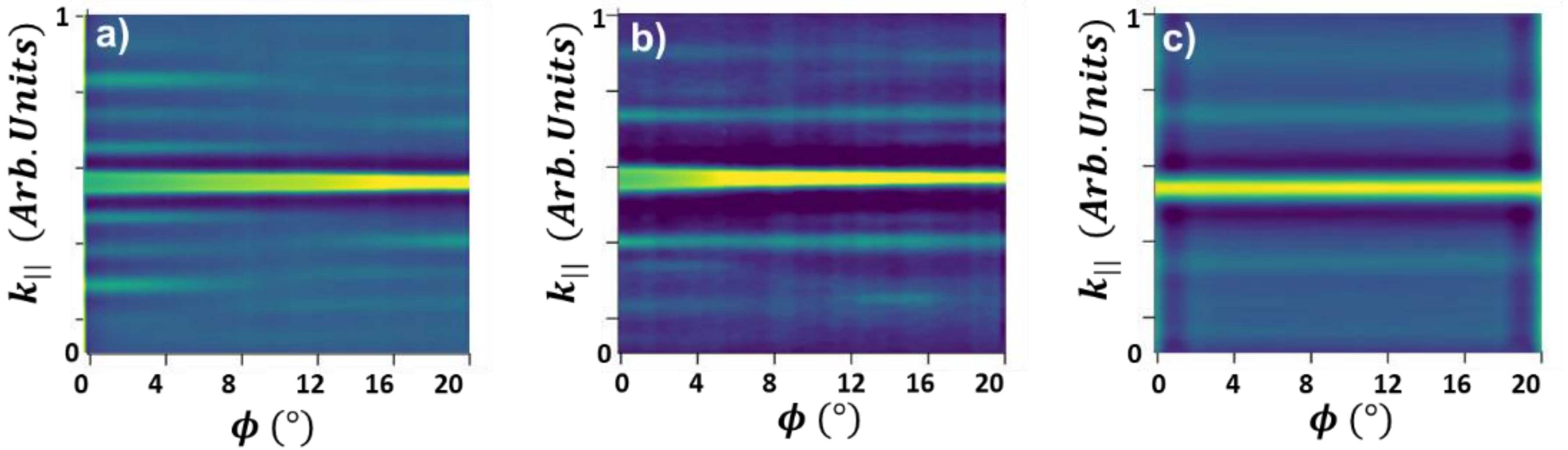
Angular dependence of RHEED of Cs_2Te grown on different substrate

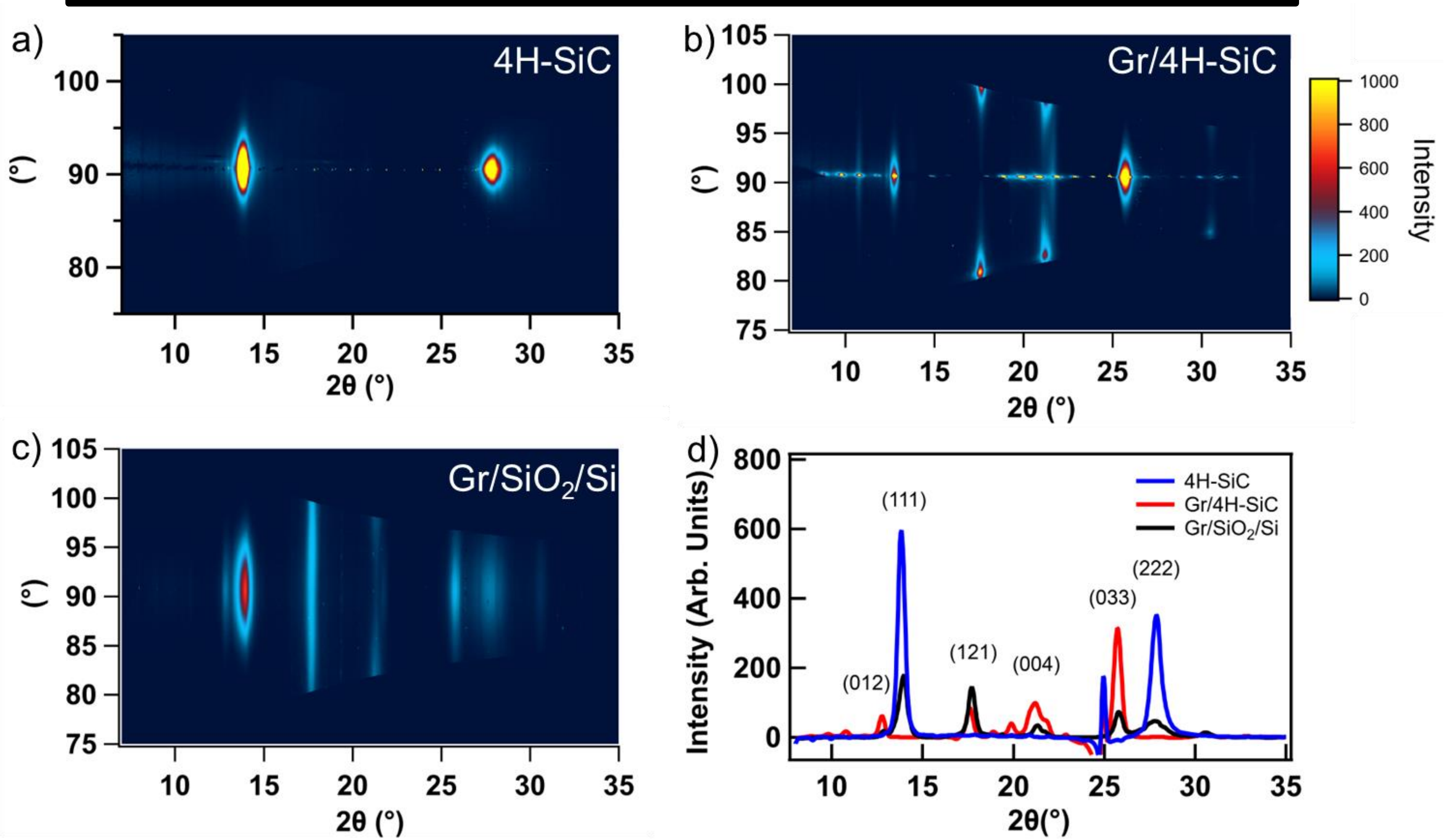


Cs_2Te/4H-SiC | Cs_2Te/Gr/4H-SiC | Cs_2Te/Gr/SiO_2/Si



- Angular dependence is observed for Cs_2Te films deposited on 4H-SiC and Gr/4H-SiC substrates implying preferred in-plane crystal orientation and epitaxial coordination with the substrates.
- No angular dependence was found for Cs_2Te deposited on the transferred graphene on SiO_2/Si implying fiber textured and polycrystalline films.

XRD of Cs_2Te grown on different substrate



- The diffraction spots of films grown on the substrates 4H-SiC and Gr/4H-SiC indicate the single crystal nature of the Cs_2Te films
- The arc-like feature and the continuous diffraction lines indicates mixture of fiber-textured and polycrystalline grains for Cs_2Te film on the transferred Gr substrate.

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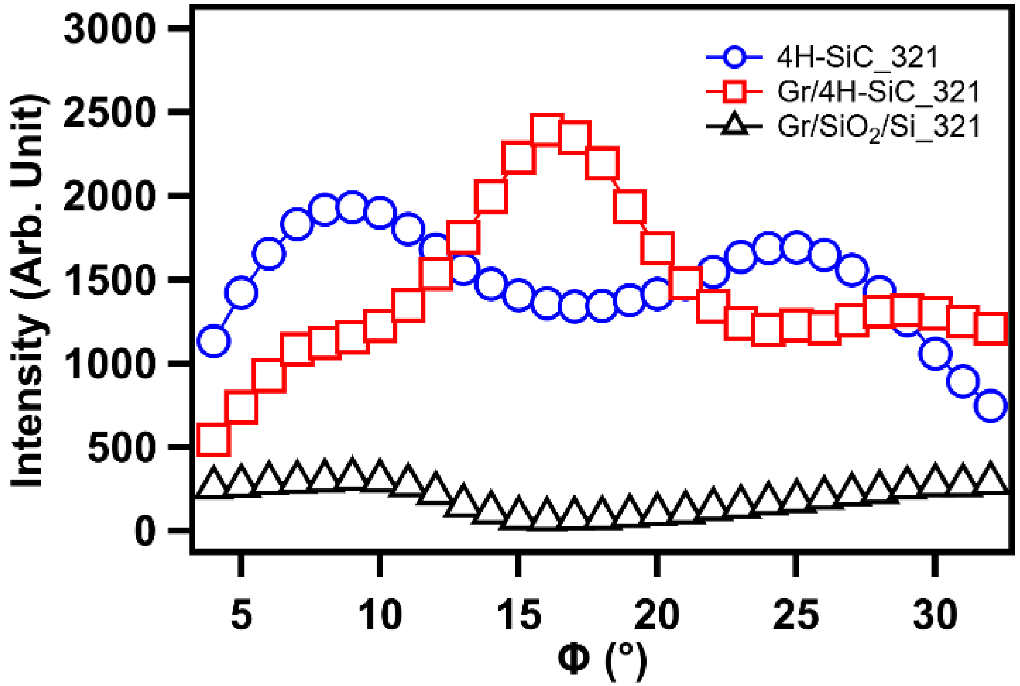
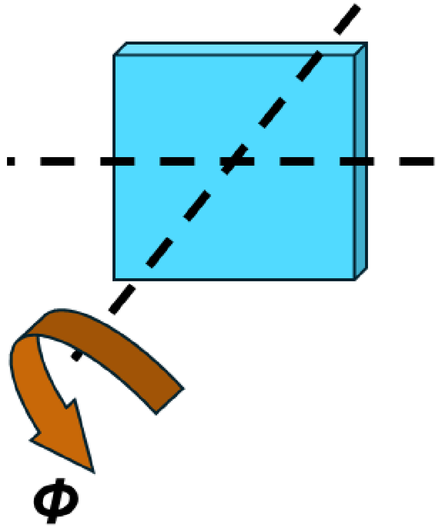
Angular dependence of XRD of Cs_2Te grown on different substrate





- The change in intensity of the Bragg reflection (321) with azimuthal rotation is observed.
- Clear angular dependence is observed for both Cs_2Te films on the 4H-SiC and the Gr/4H-SiC substrates, indicating the formation of highly ordered crystalline domains in the bulk of the cathode films.

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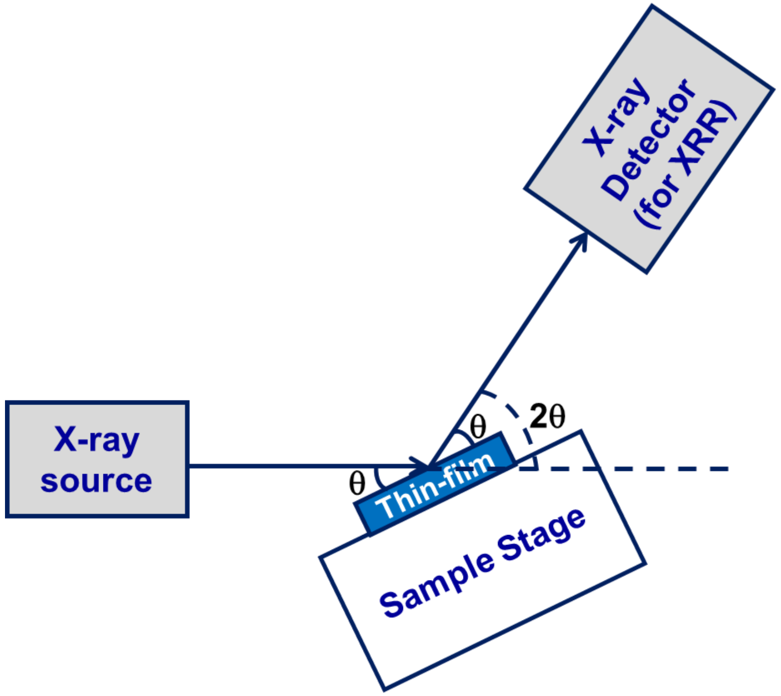
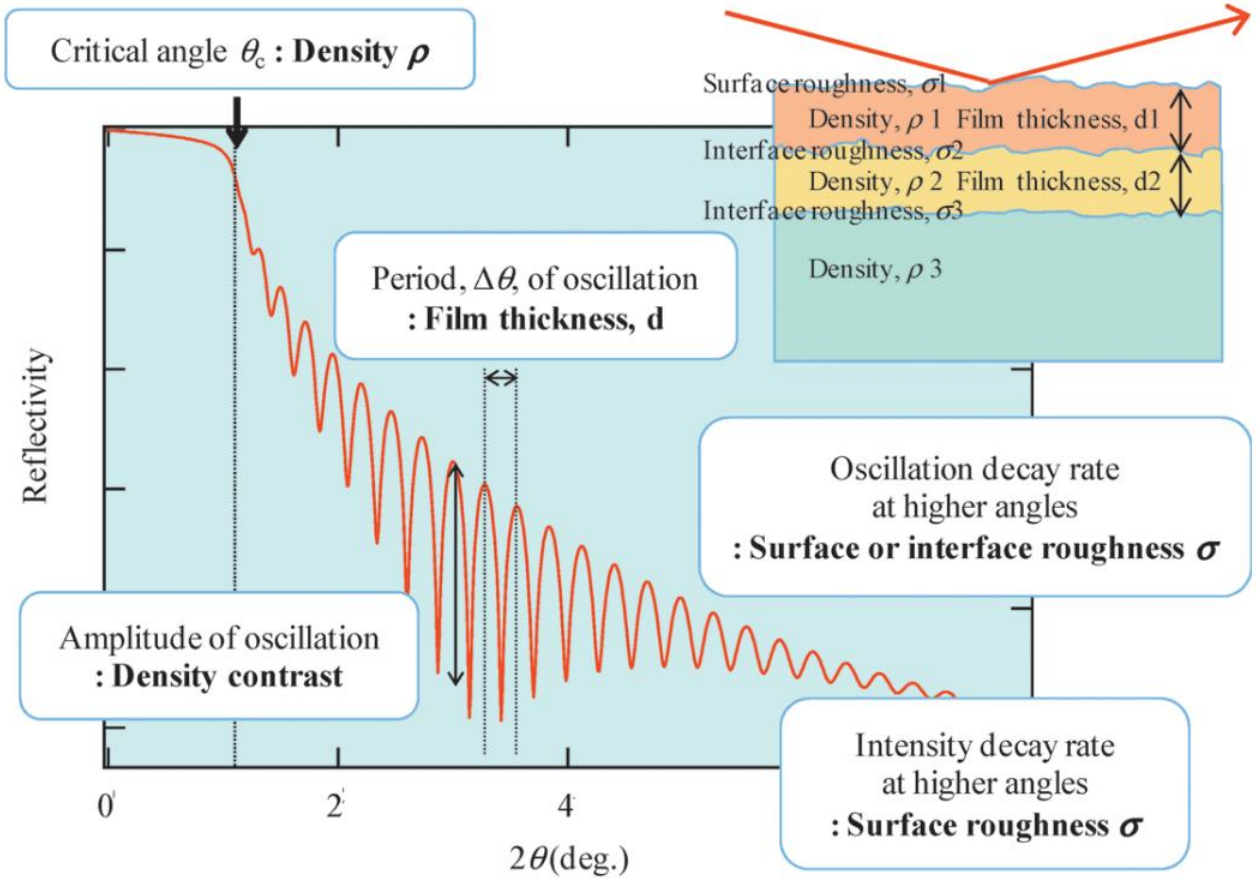
XRR of Photocathode

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]

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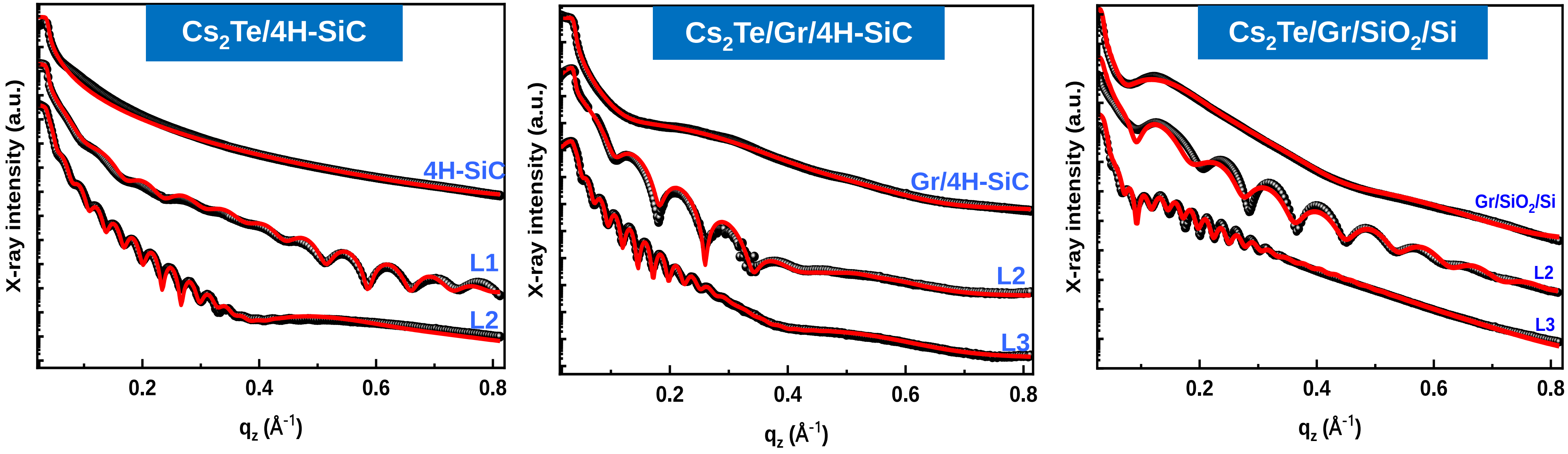
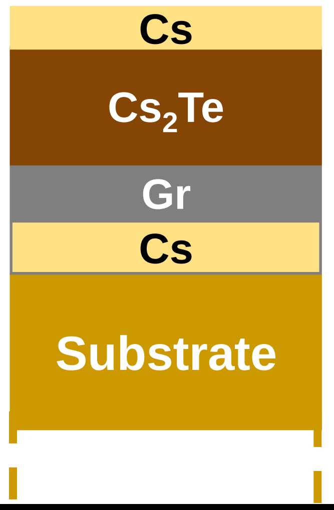
XRR & XRF of Cs_2Te on different substrate





Schematic of fitting model of Gr Substrate

Sample	Cs_2Te/4H-SiC			Cs_2Te/Gr/4H-SiC				Cs_2Te/Gr/SiO_2/Si			
	Substrate	L1	L2	Sub.	L1	L2	L3	Sub.	L1	L2	L3
Thickness (nm)	-	7.7	20.8	-	-	7.1	22.7	-	-	7.7	24.6
Roughness (nm)	0.20	0.70	0.90	0.20	-	0.6	0.8	0.45	-	0.5	0.6
Stoichiometry (Cs:Te)	-	2.03	1.98	-	2.50	2.12	1.91	-	2.87	2.22	1.94

- ❑ Smooth thin film has been prepared; roughness is less than 1 nm.
- ❑ Good fitting required excess Cs layer in the interface of Gr and 4H-SiC or SiO_2/Si and top of Cs_2Te.
- ❑ Stoichiometry of Cs_2Te obtained from XRF also suggests intercalation of Cs.

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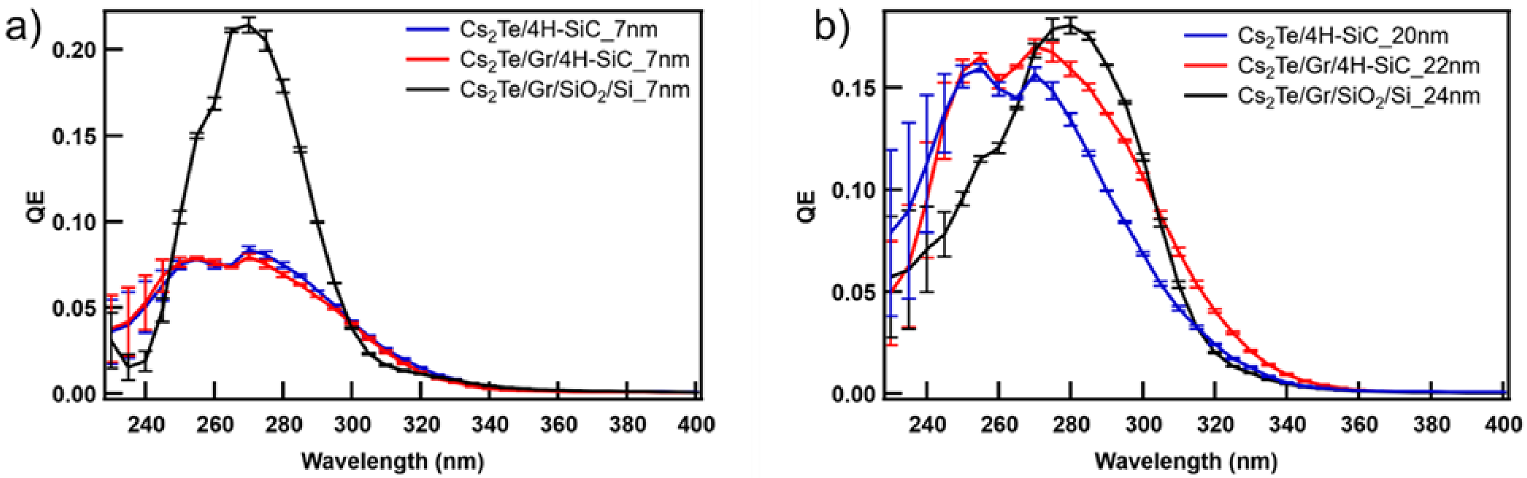
Spectral response of Cs_2Te

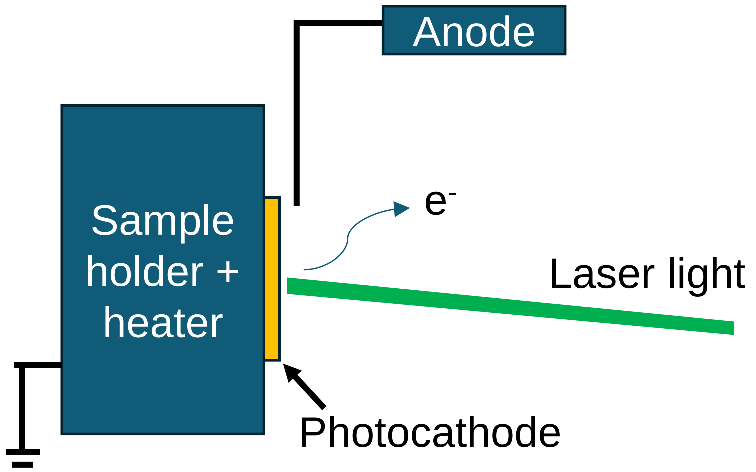
Quantum Efficiency (QE) of Photocathode

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.





Wavelength (nm)	QE (%)					
	Cs_2Te/4H-SiC		Cs_2Te/Gr/4H-SiC		Cs_2Te/Gr/SiO_2/Si	
	L1	L2	L2	L3	L2	L3
265	7.0	13.5	6.9	15.0	20.0	13.0
Peak (at 270 nm)	8.4	15.5	8.0	17.1	21.5	18.4 (at 280 nm)
Thickness (nm) [from XRR]	7.7	20.8	7.1	22.7	7.7	24.5

- ❑ QE ~7% and ~ 8% peak at 265 and 270 nm respectively obtained from ~7 nm Cs_2Te photocathode deposited on 4H-SiC and Gr/4H-SiC substrates, and that of 20% and 21.5 % peak at 265 and 270 nm from Cs_2Te/Gr/SiO_2/Si.
- ❑ QE ~15% at 265 nm and ~15.5% and 17 % at 270 nm obtained from ~20 nm Cs_2Te photocathode deposited on both 4H-SiC and Gr/4H-SiC substrates, and that of 13% at 265 nm and peak 18.5% at 280 nm from Cs_2Te/Gr/SiO_2/Si.
- ❑ The higher QE observed Cs_2Te on Gr/SiO_2/Si substrate is likely due to the optical interference arising from the SiO_2 substrate.

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Conclusions

- Successful in growing epitaxial thin film of Cs_2Te photocathode using PLD.
- Successful in growing very smooth surfaces of Cs_2Te photocathode with high crystallinity
  - XRR provides information about the roughness of <0.7 nm for 7 nm and <1 nm nm for 20 nm Cs_2Te thin film thickness.
  - RHEED also confirms epitaxial photocathode with smooth surface with small domains.
- XRF, RHEED, and XRD confirm the epitaxial growth of Cs_2Te and Stoichiometry.
- High QE has been achieved with epitaxial Cs_2Te photocathode
  - QE ~ 17% and 8% at 270 nm from 20 nm and 7 nm of Cs_2Te on 4H-SiC and Gr/4H-SiC.
  - QE ~ 18% and 21% at 270 nm from 20 nm and 7 nm of Cs_2Te on Gr/SiO_2/Si
  - QE of the co-dep cathode reached 19% at 266 nm from 100 nm Cs_2Te, while the sequentially grown cathode yields a QE of 8.5% at the same wavelength [Gaowei *et al* (2019)]

Future Plans

- Achieve high QE with deposition high enough film thickness.
- Measure the emittance of the epitaxial Cs_2Te photocathodes.

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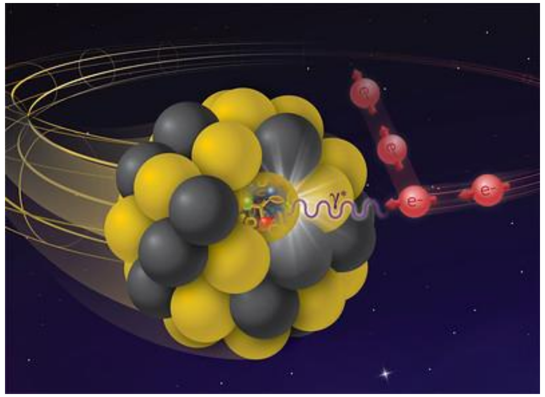
Epitaxial Growth of K_2CsSb

- BNL group is interested 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 maintain hadron emittance by using hadron cooling.



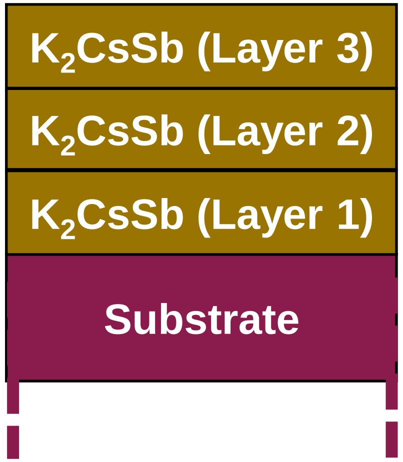
Electron beam for EIC Hadron Cooling

- High average current (100 mA)
- High bunch charge (up to 2 nC)
- Long lifetime (> 3 days)
- Low emittance

- Bi-alkali antimonide (K_2CsSb) photocathodes are selected to meeting these requirements, it is also the electron source material currently used at LEReC and CeC

PLD and thermal evaporation-assisted epitaxial growth at BNL

- Sb deposited using PLD and thermal evaporation.
- Cs and K were deposited using effusion cells.
- Layer 1: K_2CsSb on substrate, for structural study about 5 nm film thickness to confirm epitaxial growth.
- Layer 2: Growth of another layer of K_2CsSb to check the continuity of epitaxial growth.
- Layer 3: Growth of photocathode to achieve high QE with epitaxial growth.



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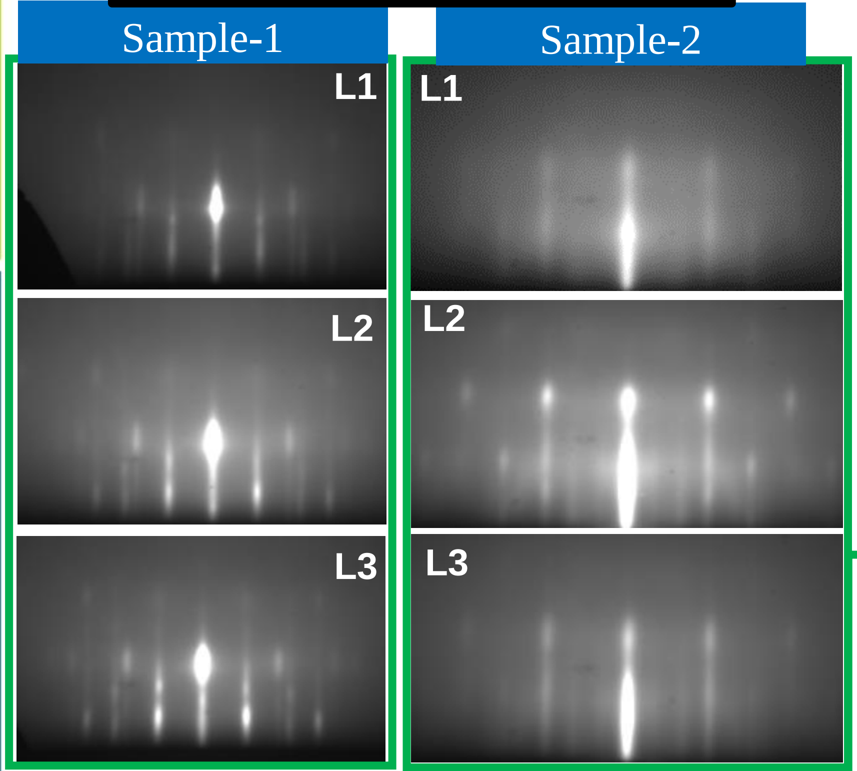
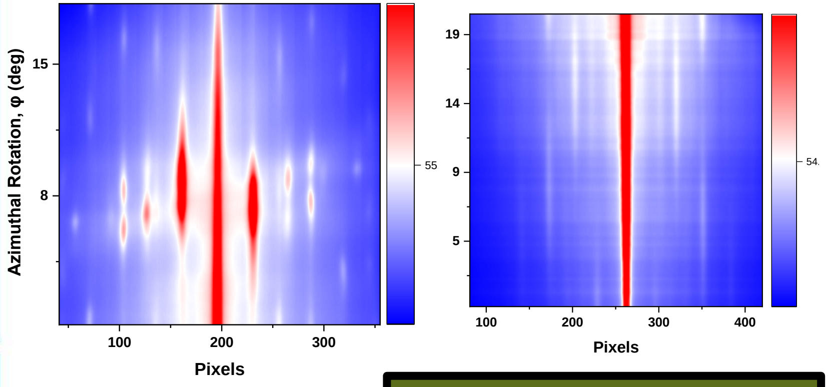
RHEED of K_2CsSb

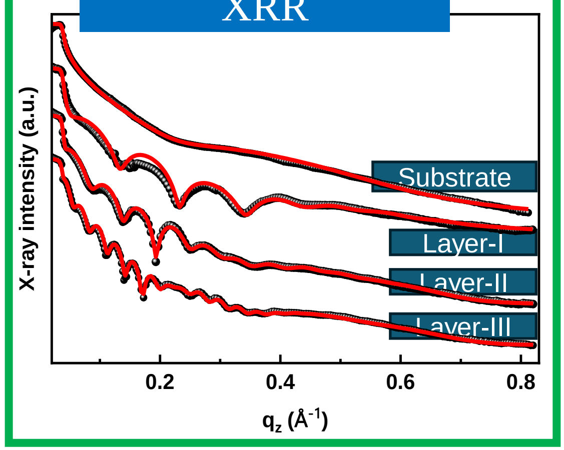


Azimuthal angular dependence

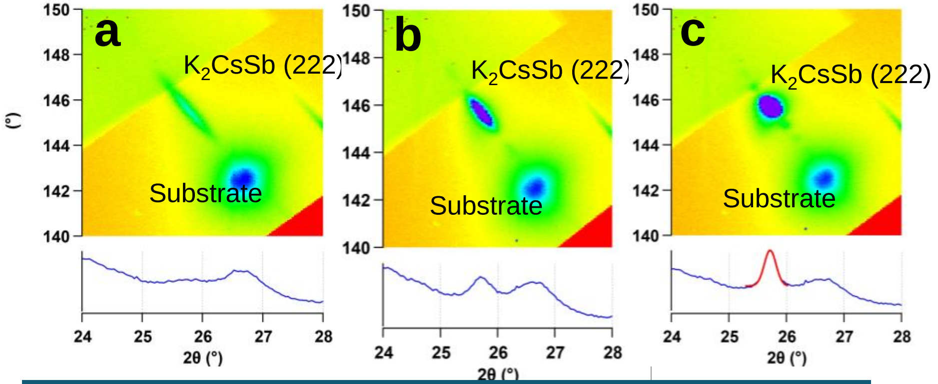


- Successful in growing very smooth surfaces of K_2CsSb photocathode with high crystallinity.
- XRR provides roughness of 0.4 nm for 5 nm thin layer and 0.8 nm for 20 nm film thickness.
- Compared to sequential growth (roughness 1.3 nm for 17 nm film [Ding *et al* (2017)]), epitaxial growth provide promising results.
- RHEED streaks confirms epitaxial photocathode with smooth surface with small domains.
- XRF, RHEED, and XRD confirm the K_2CsSb Stoichiometry with epitaxial growth.
- Green laser: QE obtained > 9%. 3.2% QE from 5 nm thin film.
- 450 nm: QE obtained > 21%. 8.9 % QE from 5 nm thin film.

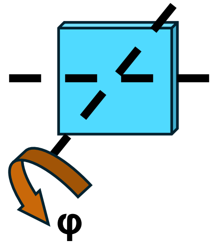
XRR



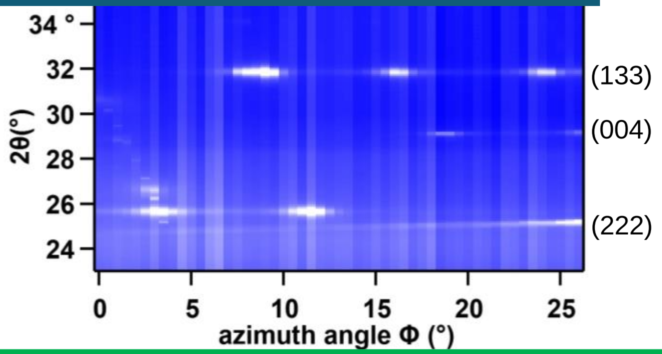
XRD for (a) K_2CsSb (L1), (b) K_2CsSb (L2), (c) K_2CsSb (L3)



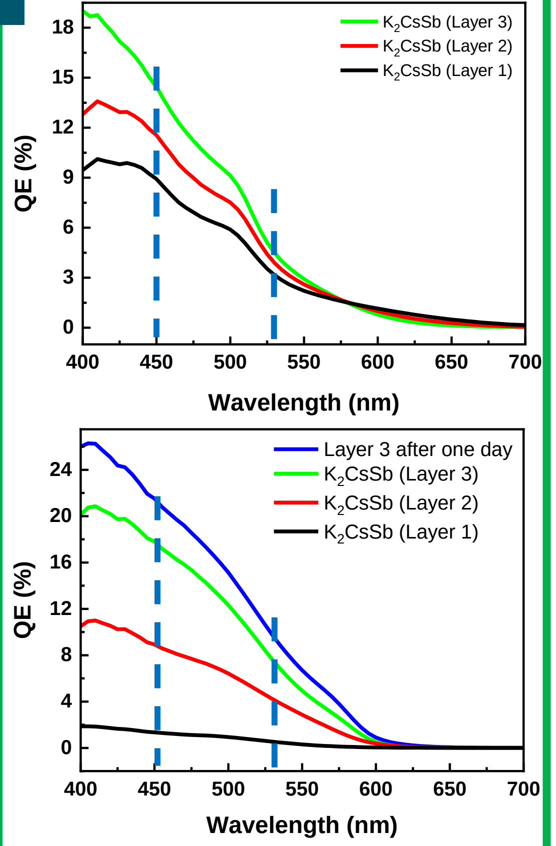
Azimuthal angular dependence of XRD of K_2CsSb



Schematic of azimuthal sample rotation



Spectral response



XRF of K_2CsSb (Layer 3)

Cs/Sb: 1.29; K/Sb:1.88

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Cornell University

Chad Pennington, Elena Maria Echeverria Mora, Jared Maxson

Arizona State University

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Leiden Institute of Physics

Molen S.J. Van der, Guido Stam

University of Vienna

Thomas Juffmann

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Thank You

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