In-situ X-ray Reflectivity Analysis of Alkali Antimonide photocathodes

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Outline

- Introduction of alkali antimonide photocathodes
- Basics of X-ray reflectivity
- Experimental details and results of a series of K_2CsSb, Cs_3Sb photocathode
  - Sequential evaporation
  - co-evaporation
  - Sputtering deposition
- Summary

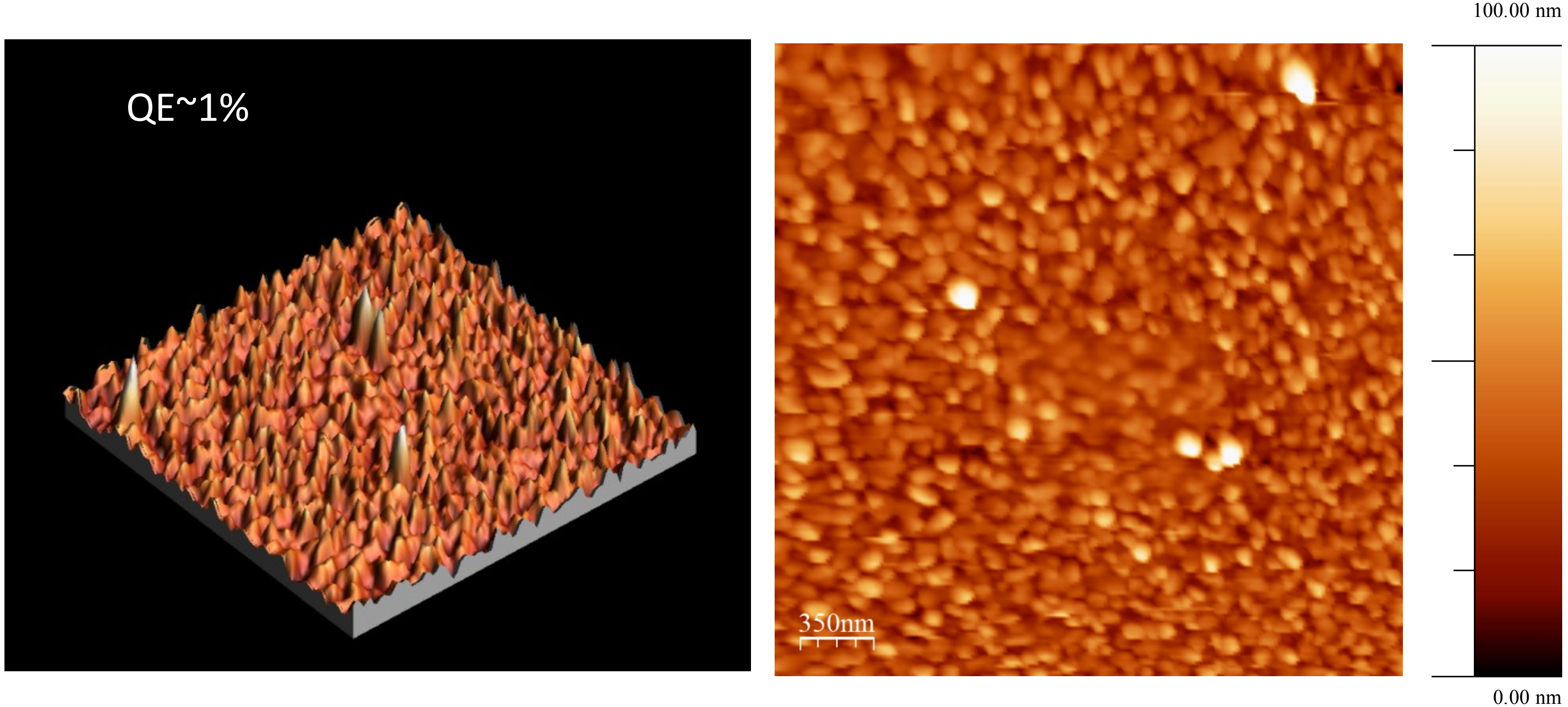
Introduction

- Alkali Antimonide (K_2CsSb, Cs_3Sb, …) – excellent photocathode candidate for accelerator
  - High quantum efficiency
  - Low emittance
  - Good lifetime
  - Fast response

Cathode type	Cathode	Typical wavelength & energy, λ_{opt} (nm), (eV)	Quantum efficiency (electrons per photon)	Vacuum for 1000 h (Torr)	Gap energy+ electron affinity, E_G+E_A (eV)	Thermal emittance (microns/ mm(rms))	
						Eq. (7)	Expt.
PEA: mono-alkali	Cs_2Te	211, 5.88	0.1	10^{-9}	3.5 [42]	1.2	0.5 ± 0.1 [35]
		264, 4.70	–	–	"	0.9	0.7 ± 0.1 [35]
		262, 4.73	–	–	"	0.9	1.2 ± 0.1 [43]
	Cs_3Sb	432, 2.87	0.15	?	1.6+0.45 [42]	0.7	?
⋮							
PEA: multi-alkali	Na_2KSb	330, 3.76	0.1	10^{-10}	1+1 [42]	1.1	?
	$(Cs)Na_3KSb$	390, 3.18	0.2	10^{-10}	1+0.55 [42]	1.5	?
	K_2CsSb	543, 2.28	0.1	10^{-10}	1+1.1 [42]	0.4	?
	$K_2CsSb(O)$	543, 2.28	0.1	10^{-10}	$1+<1.1$ [42]	~0.4	?

D.H. Dowell et al. / Nuclear Instruments and Methods in Physics Research A 622 (2010) 685–697

Introduction – Roughness From Traditional Recipe



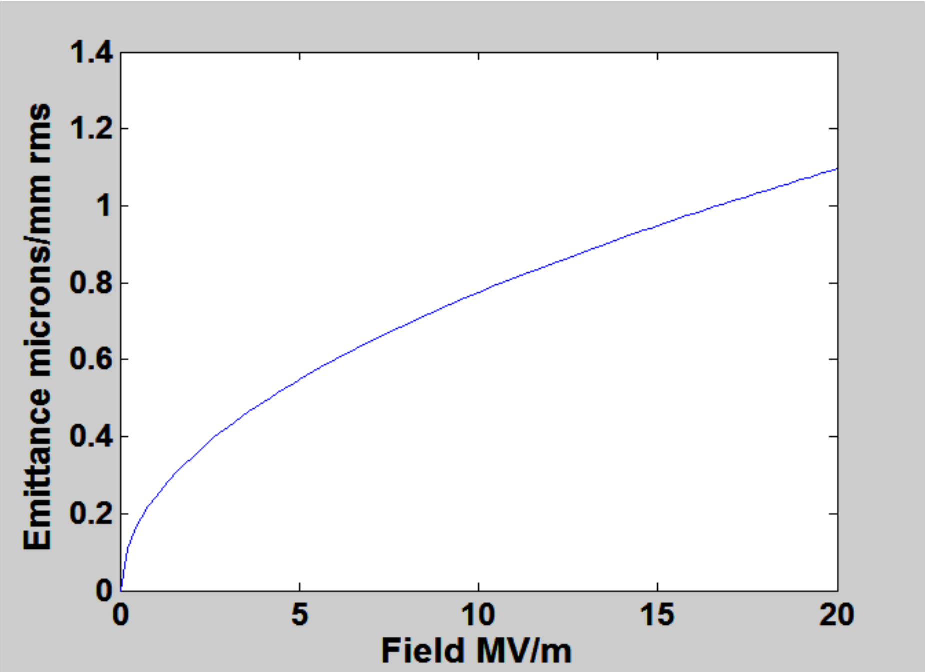
Previous study shows rms roughness of 25nm over 100 nm spatial period on a K_2CsSb photocathode grown by sequential evaporation.

S. G. Schubert, et al, APL Mater. **1**, 032119 (2013)

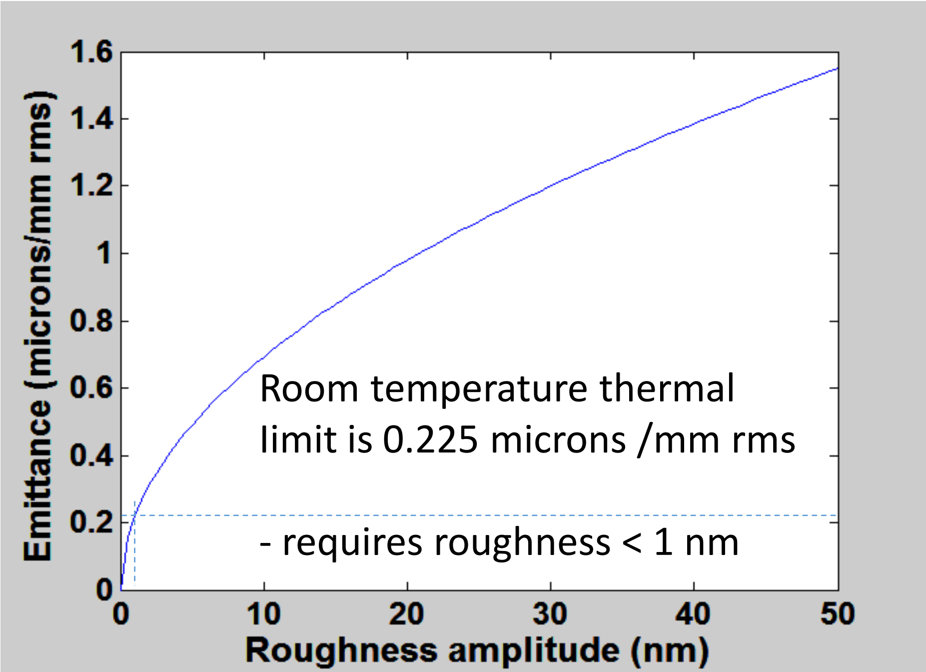
Introduction – Effect of roughness on thermal emittance

$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2}{2m_0 c^2 \lambda}Ee}$$

D. Xiang et al. Proceedings of PAC07, Albuquerque, New Mexico, USA

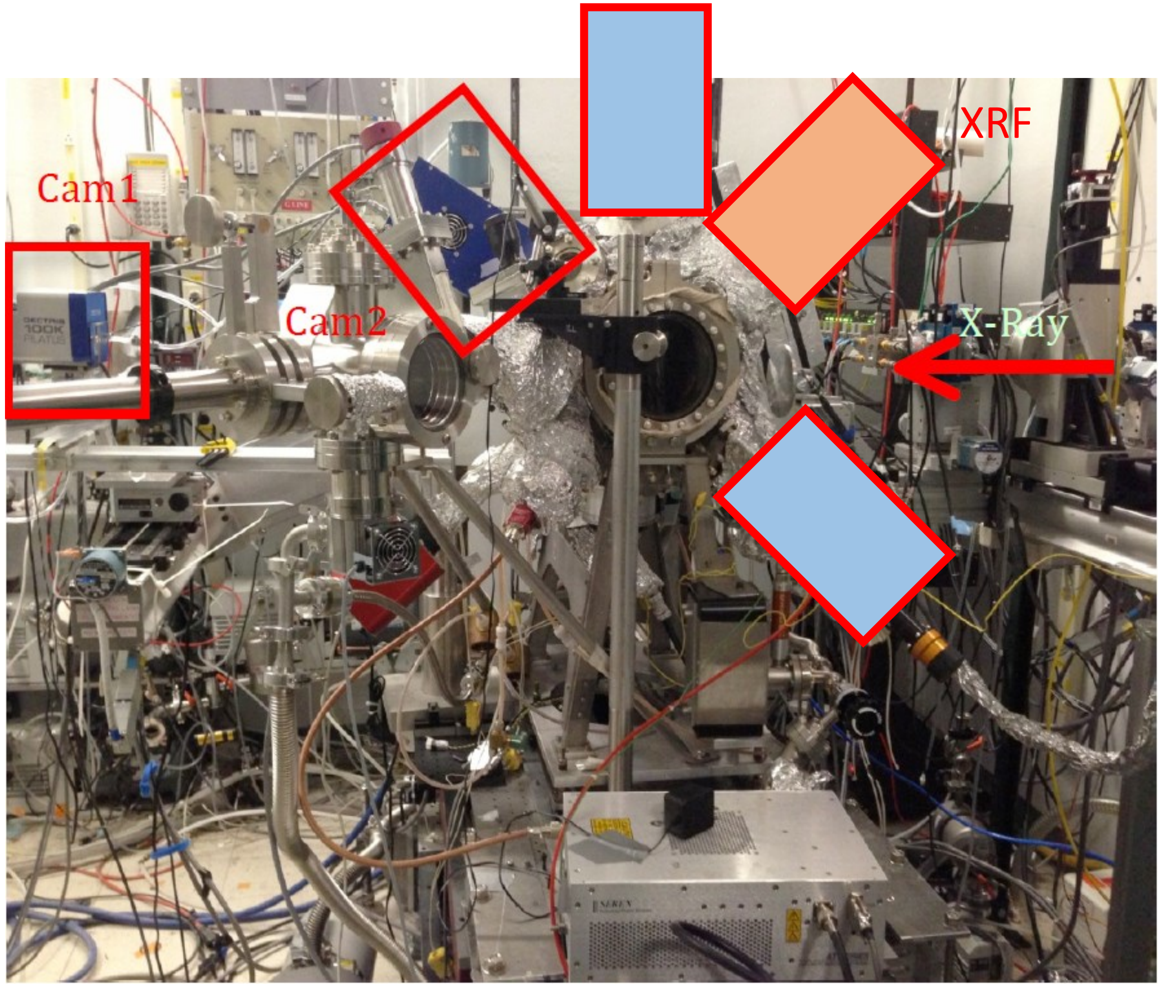


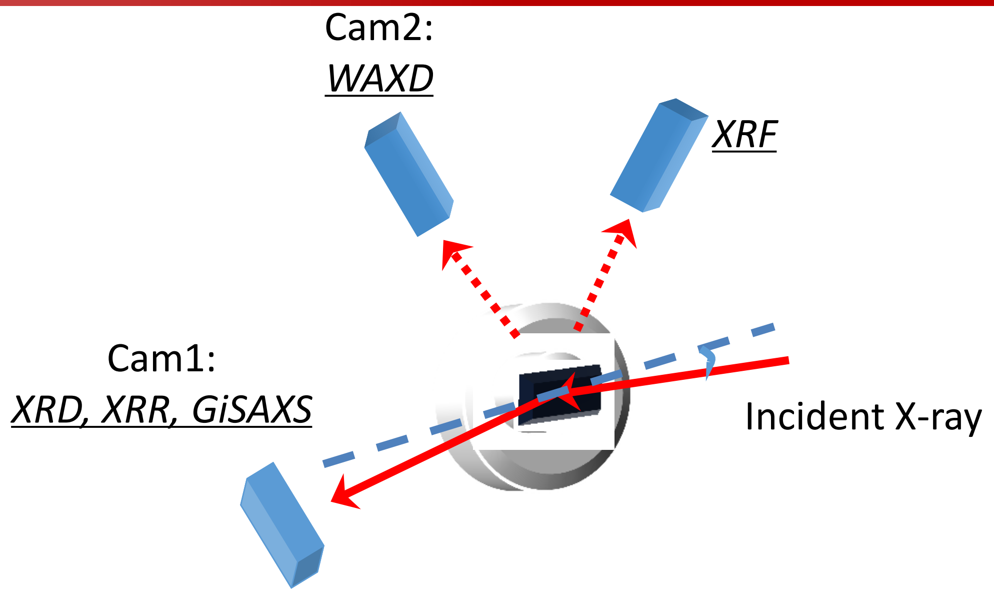
Field dependent emittance growth: 20 nm amplitude, 80 nm period

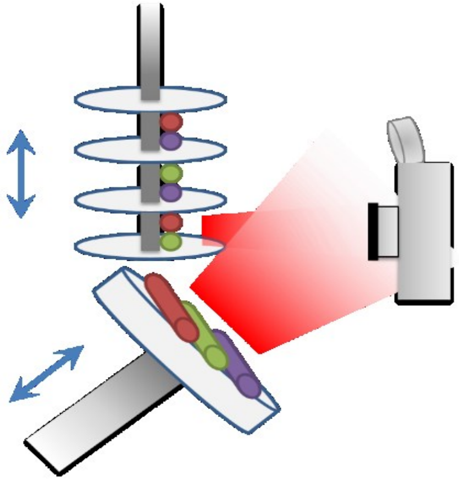


Emittance growth at 20 MV/m, period 4 x amplitude.

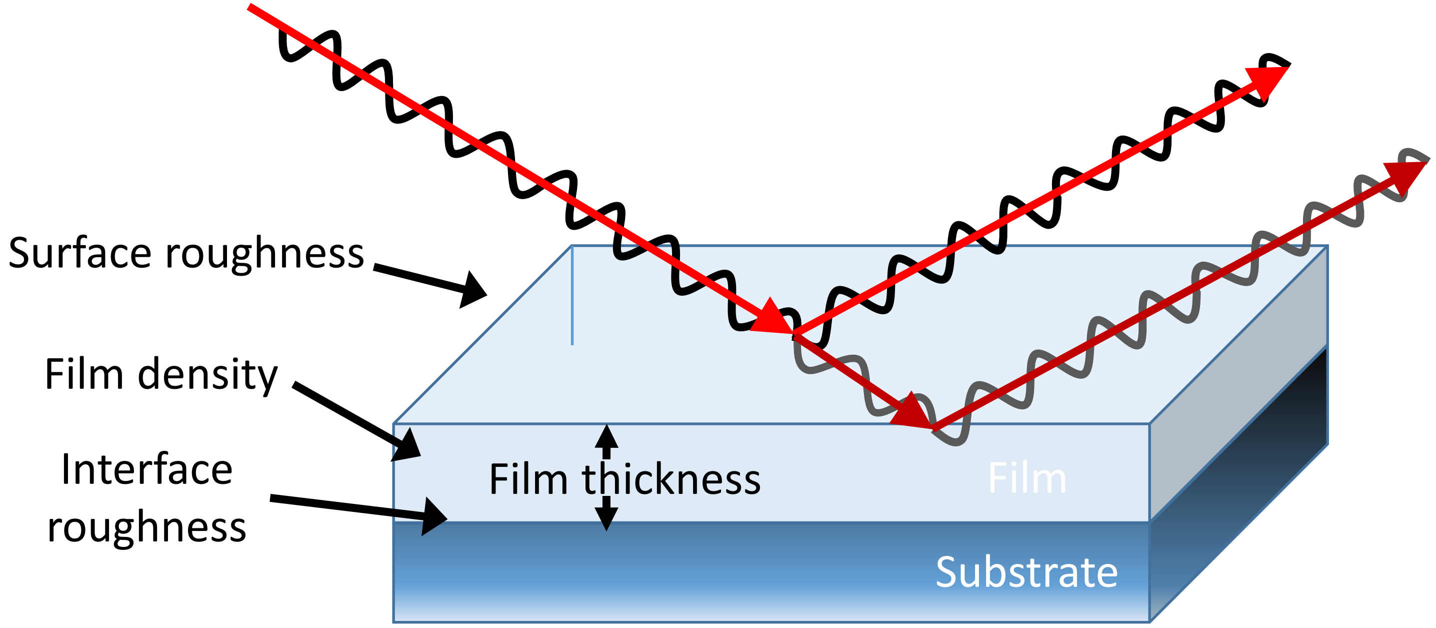
Introduction – In-situ Growth System

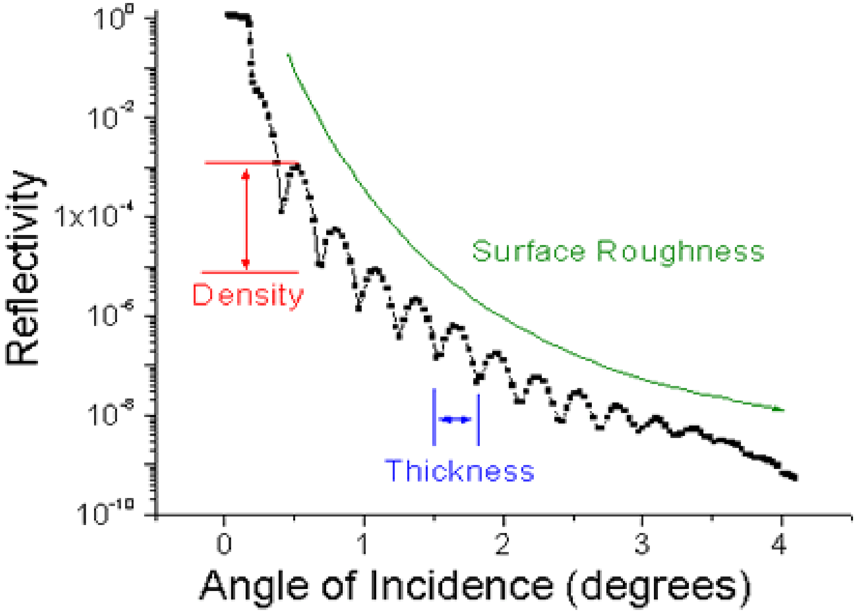






Basics of X-ray Reflectivity

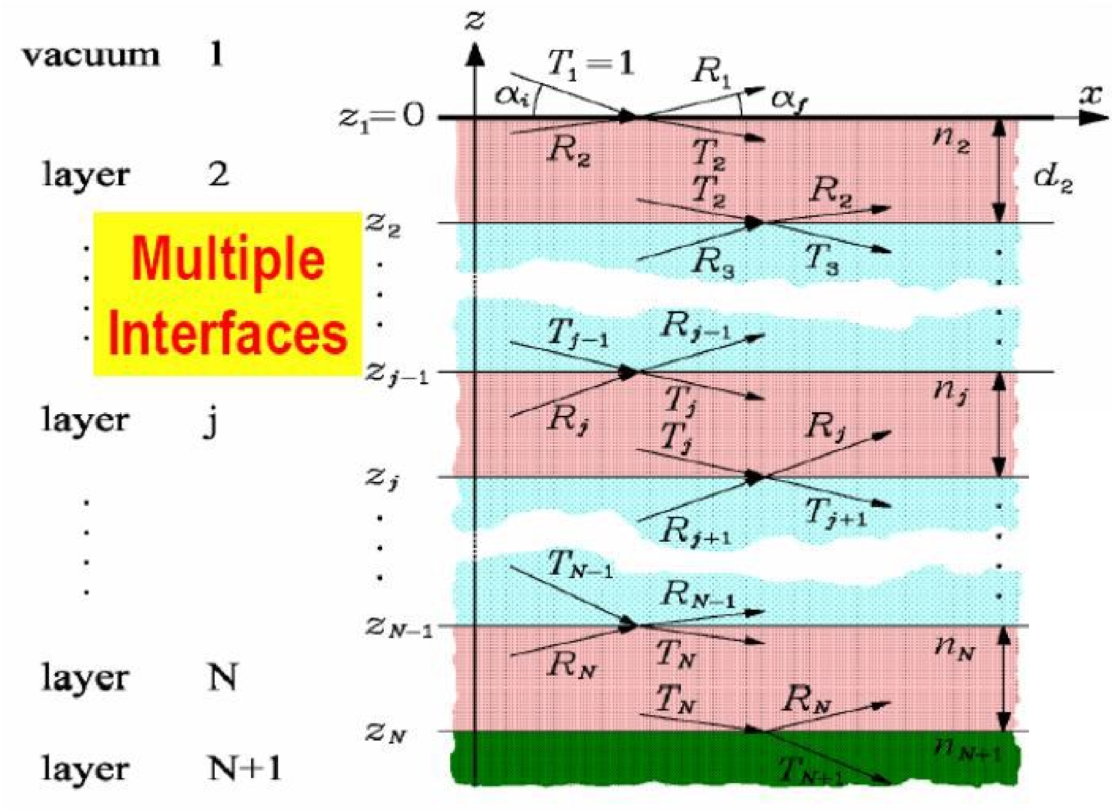




X-ray reflectivity: a fast and non-destructive technique to characterize thin film properties

Basics of X-ray Reflectivity

- Parratt recursion, Parratt, 1954.



Multilayer thin film → stratified medium

For *j*th layer:

$$r_{j,j+1} = \frac{q_{z,j} - q_{z,j+1}}{q_{z,j} + q_{z,j+1}} \Rightarrow$$

$$r = \sum_{j=0}^{n} r_{j,j+1} e^{iq_z \sum_{m=0}^{j} d_m}$$

- Nevot-Croce model for roughness calculation

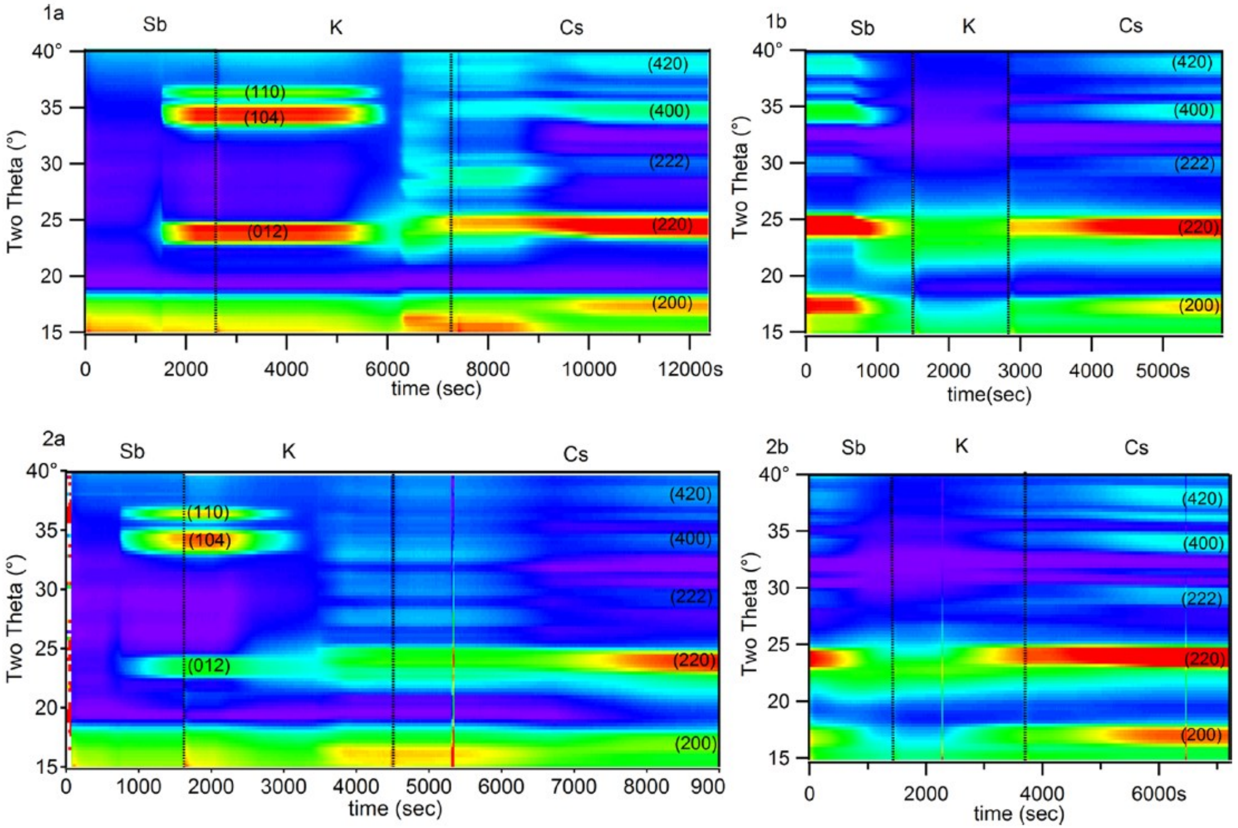
$$r'_{j,j+1} = r_{j,j+1} e^{-2k_{z,j}k_{z,j+1}\sigma_j^2}$$

$\Rightarrow$ *Intensity decreases exponentially with* σ_j^2

Parratt, L. G. (1954). Phys. Rev. 95, 359.

X-ray and Neutron Reflectivity, J. Daillant, A. Gibaud

Introduction – Previous Studies



Crystal structure evolution of K_2CsSb photocathode grown by traditional recipe

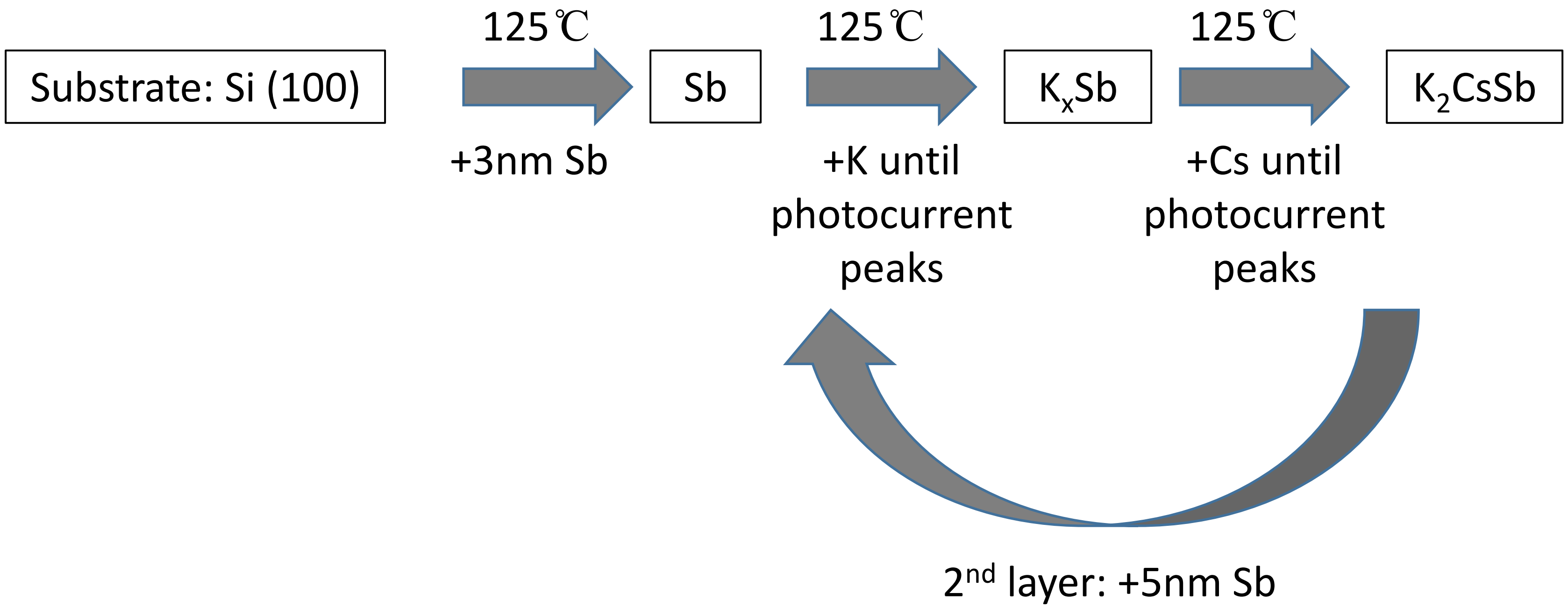
- Critical Sb-crystallization thickness: 4nm
- Surface roughening occurs due to transition from crystalline Sb (Sub nm)to crystalline K_3Sb (a few nm)

Ruiz-Osés *et al.* APL Mater. **2**, 121101 (2014)

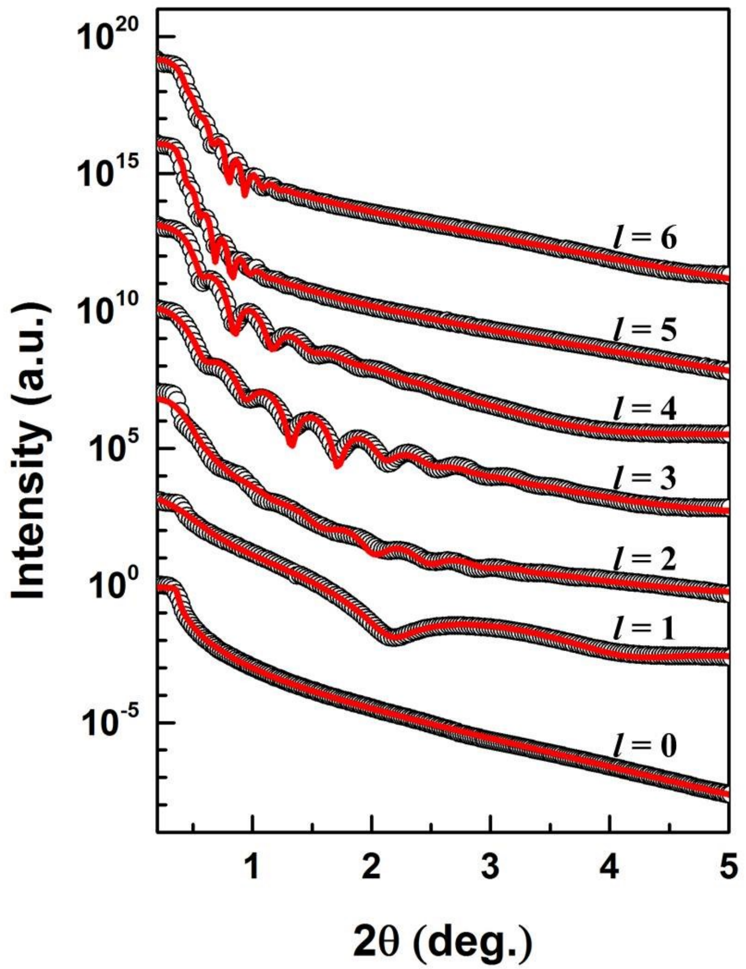
Experimental details – Sequential deposition

- Sequential deposition – *Towards a smoother surface*
  - Idea: to prevent Sb from crystallizing.
  - Critical thickness for transition from amorphous Sb to crystalline Sb is 4nm !

    (M. Ruiz-Osés et al., APL Mat. 2, 121101 (2014))
  - Procedures:



Results – Sequential deposition

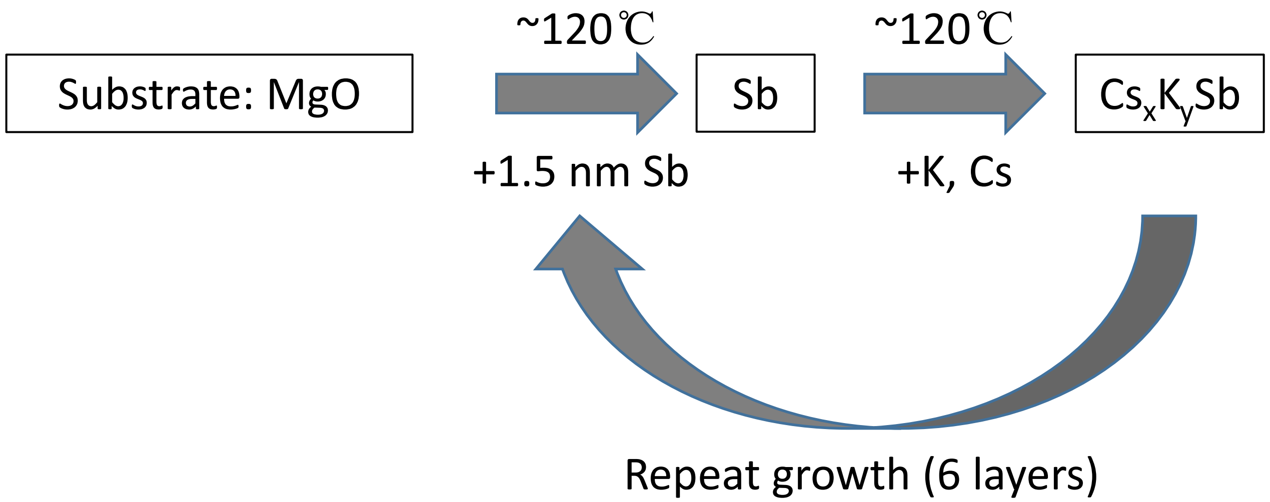


Layer ID	Recipe	QE after deposition	Total film thickness (XRR)	Roughness (XRR)
l = 6	Cs-K-Sb-Cs-K-Sb/Si	4.9%	469 Å	32.0 Å
l = 5	K-Sb-Cs-K-Sb/Si	0.88%	449 Å	36.0 Å
l = 4	Sb-Cs-K-Sb/Si	-	200 Å	21.3 Å
l = 3	Cs-K-Sb/Si	3.1%	174 Å	13.2 Å
l = 2	K-Sb/Si	0.16%	141 Å	10.5 Å
l = 1	Sb/Si	-	35 Å	2.9 Å
l = 0	Si substrate	-	-	3.1 Å

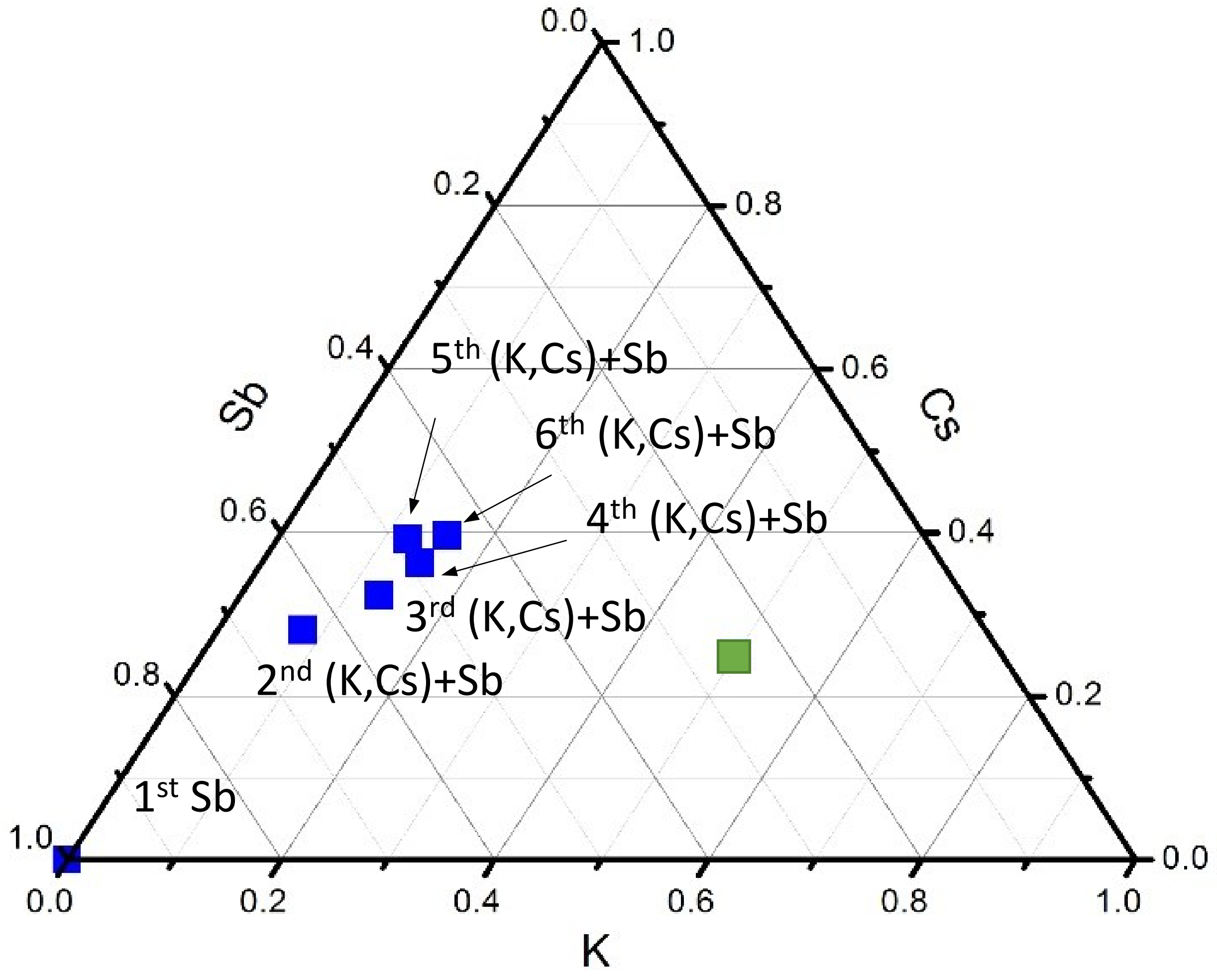
XRR simulation results of sequentially evaporated cathode

Experimental details – K/Cs co-evaporation

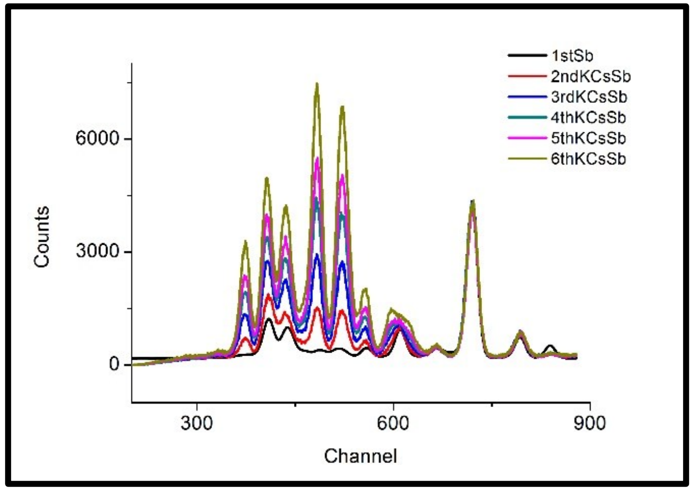
- K+Cs co-evaporation
  - Co-evaporate alkali materials on 1.5 nm Sb layer.
  - Using MgO as the substrate.
  - Procedures:



Results and Analysis



Final cathode chemical formula: $K_{0.35}Cs_{0.89}Sb$

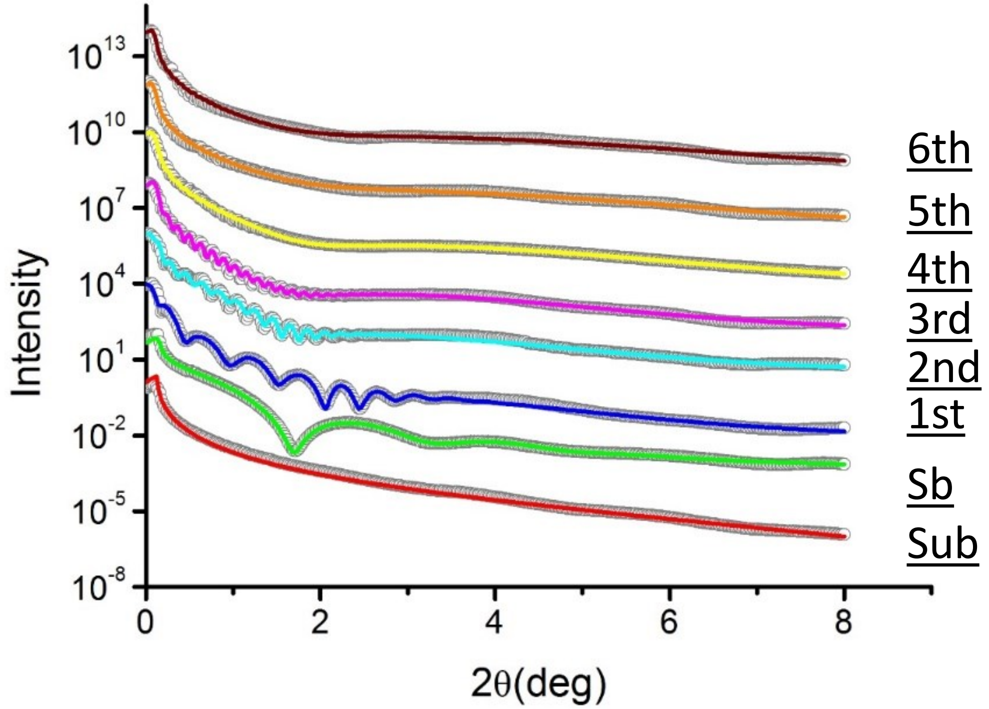


XRF analysis results of co-dep sample.

→ Calculated stoichiometry shows Sb excess, K-deficient

Chemical formula: K

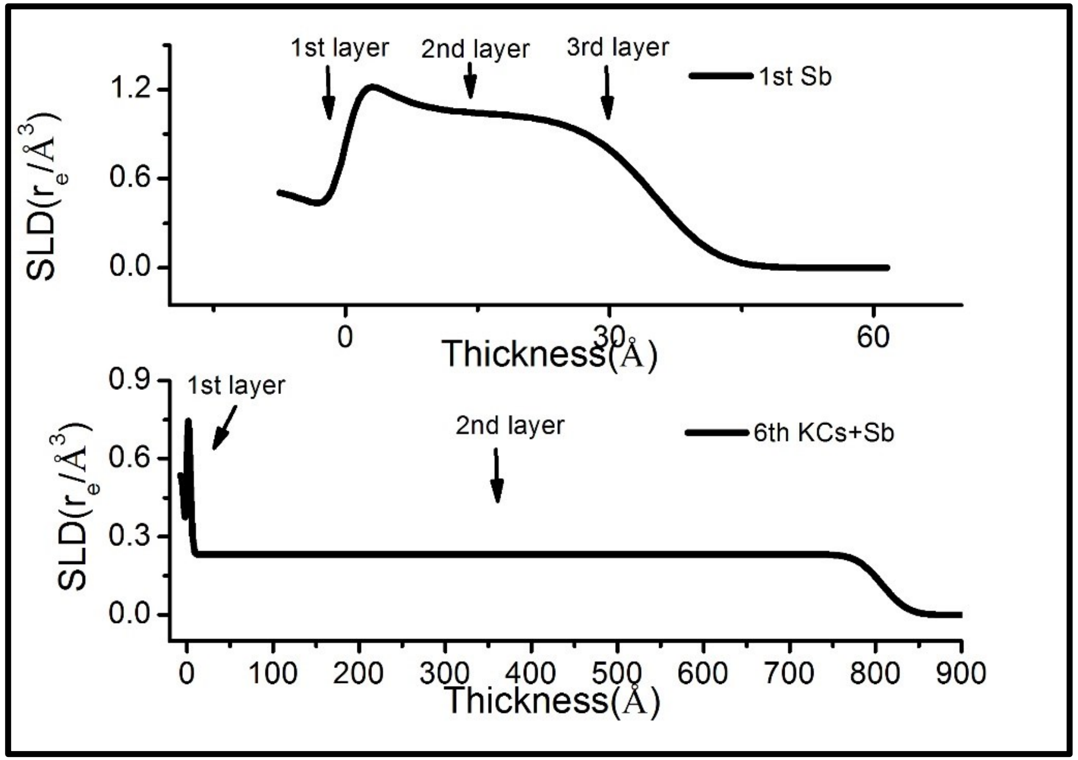
Results and Analysis



layer	Roughness (Å)	Thickness(Å)	QE
6th (K,Cs)+Sb	23.6 (-3.9, 1.4)	806.7 (-25.3, 98.9)	4.5%
5th (K,Cs)+Sb	24.9 (-17.2, 1.0)	725.5 (-46.5, 24.6)	4.9%
4th (K,Cs)+Sb	14.5 (-2.1, 0.40)	609.3 (-70.5, 9.5)	3.7%
3rd (K,Cs)+Sb	9.92 (-1.5, 1.5)	489.09 (-6.8, 18.7)	4.2%
2nd (K,Cs)+Sb	9.73 (-0.30, 0.86)	334.3 (-1.8, 2.1)	1.7%
1st (K,Cs)+Sb	9.22 (-2.9, 0.78)	159.3 (-1.6, 0.9)	1.2%
Sb	5.2 (-0.11, 0.35)	36.74 (-0.26, 0.13)	
Substrate (MgO)	1.5 (-, -)	--	--

XRR simulation result of K/Cs co-dep sample.
Colored solid lines: simulation; Open circle: measured data. Error noted is 5% of change in logarithm FOM function

Results and Analysis



Scattering length density (SLD) vs. thickness

$$\text{Re (SLD)} = 2\pi\delta/\lambda^2 = r_e\rho_e$$

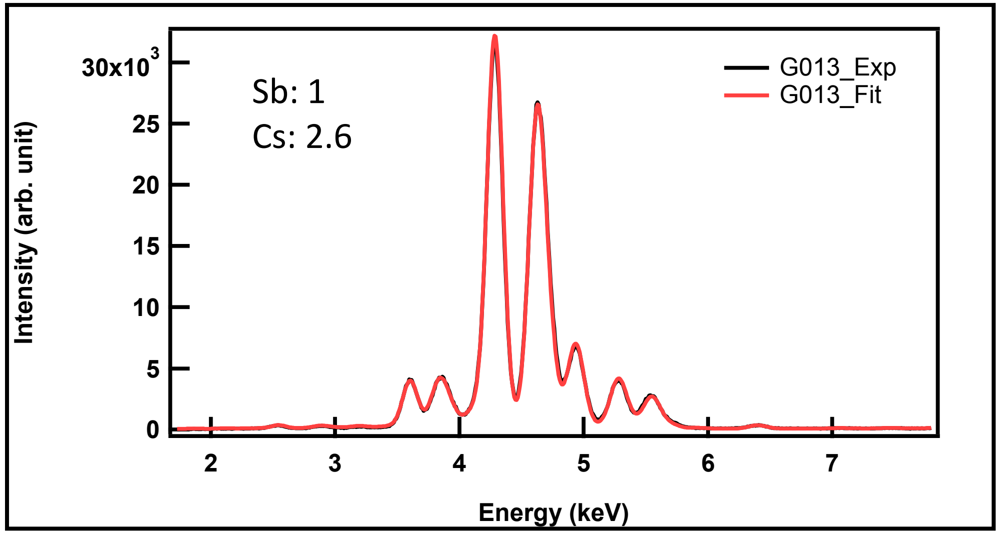
→ Simulated electron density decreases as cathode grows thicker.

→ Evaporated photocathode might be porous

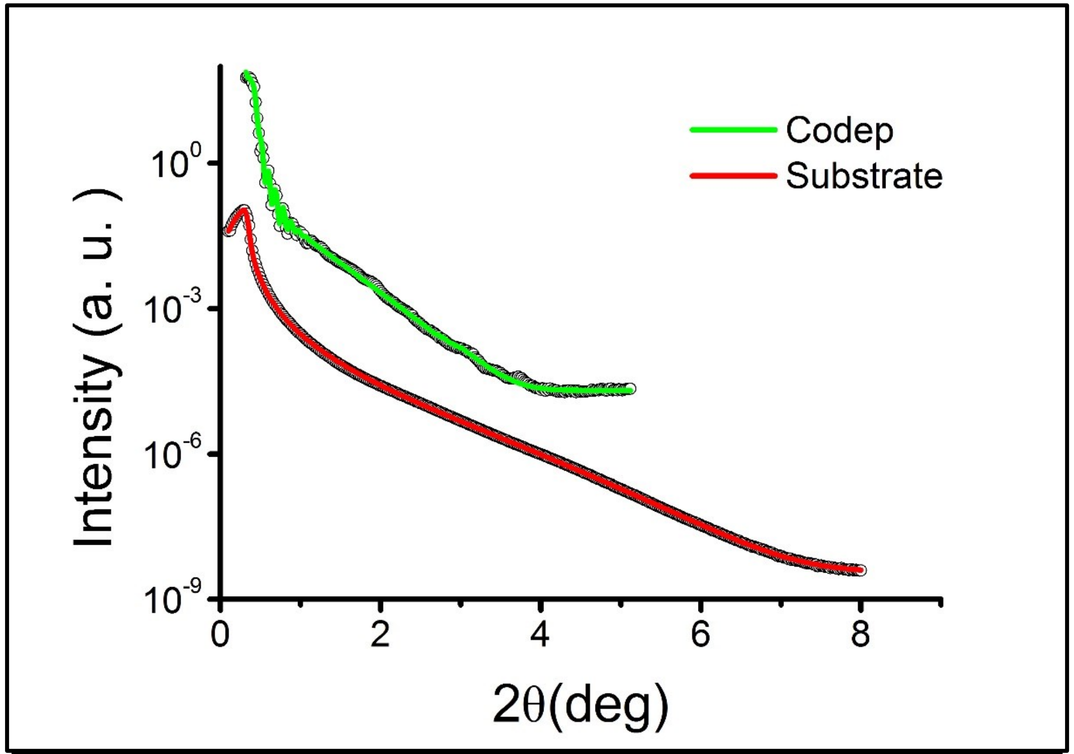
Results and Analysis – Cs/Sb co-evaporation

Recent:

- Co-evaporation of Cs/Sb @ 90℃ on Si (100)



- XRF fitted result, chemical formula $Cs_{2.6}Sb$
- QE @ 532 nm: ~4%



Layer	Thickness (Å)	Roughness (Å)
Co-dep Cs/Sb	611.2	30.6
SiO2	5.0	3.32
Substrate (Si)	--	3.75

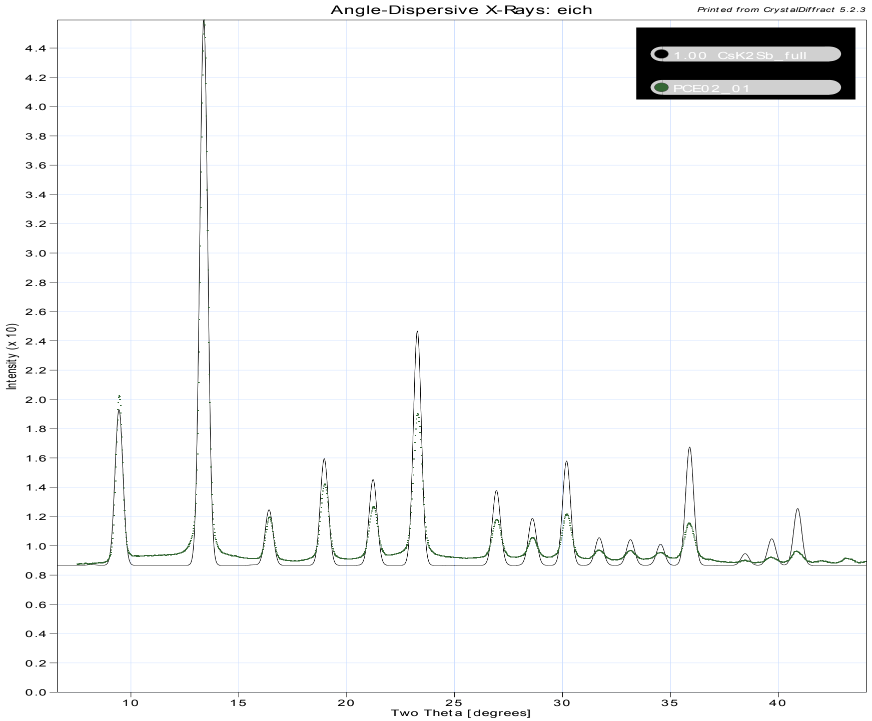
- XRR fitted result
- Not uniform in electron density

Experimental details – Sputter deposition

- <u>Towards a smoother photocathode with high QE – Sputter all materials at the same time.</u>

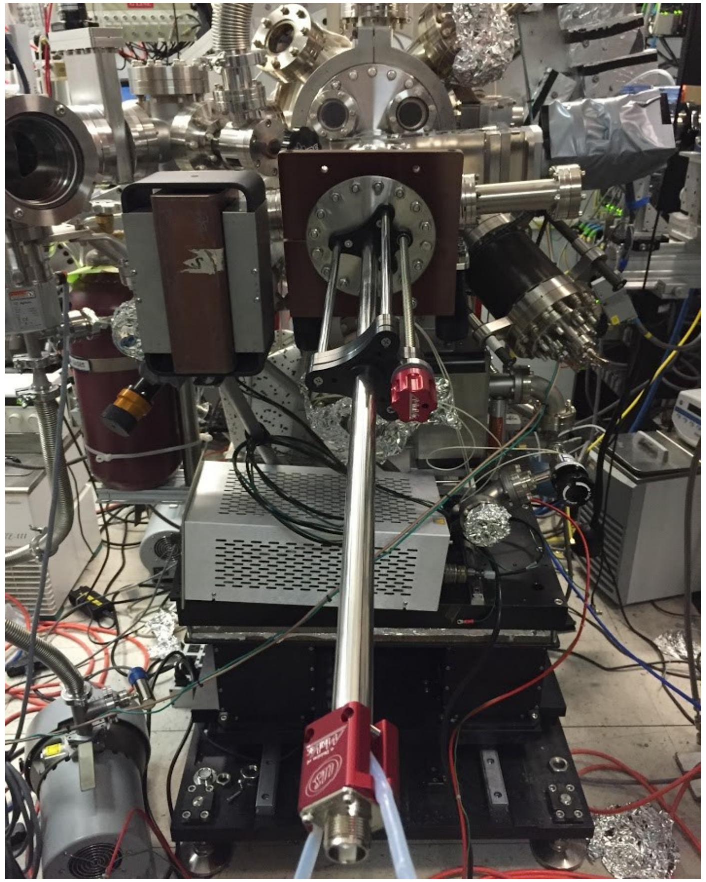
BNL Patent Hold

- Cooperation with Radiation Monitoring Devices



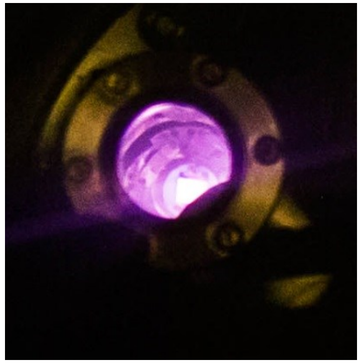
K_2CsSb target

Experimental details – Sputter deposition



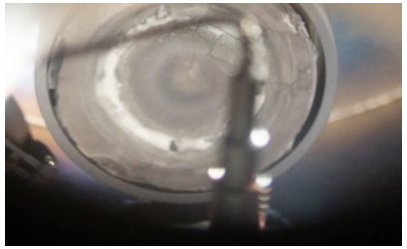
In-situ growth chamber with sputter system installed at G3, CHESS

BNL Patent Hold



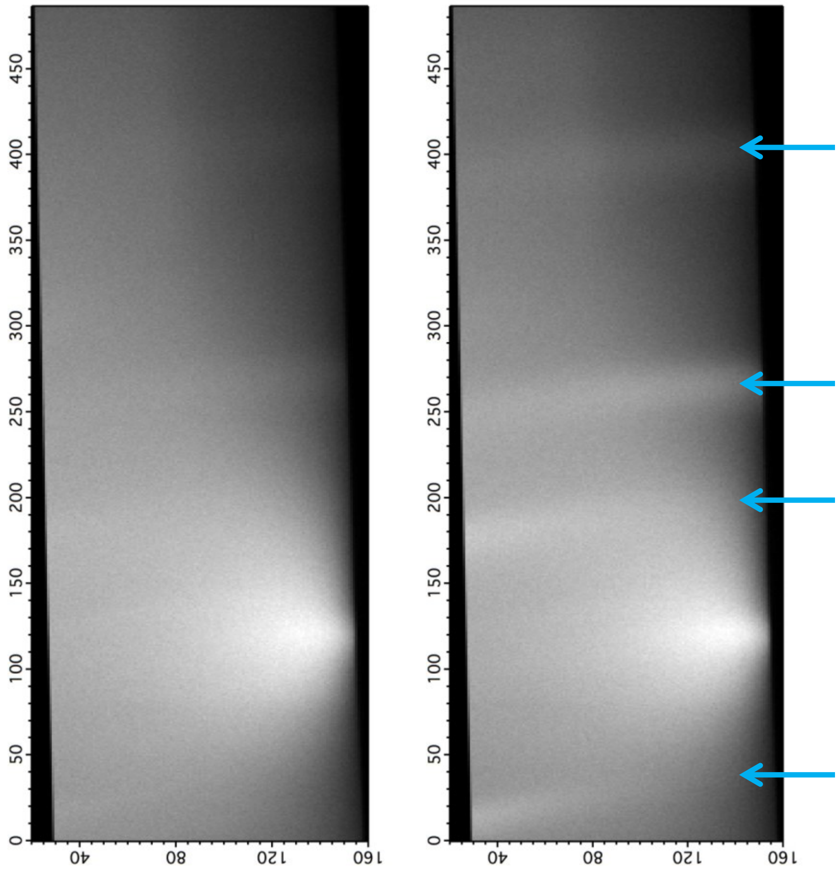
- Sputter conditions
  - 20 mTorr of Argon atmosphere.
  - Substrate temperature: ~120℃
- Procedures
  - 20W to pre-sputter for 2-3 min.
  - 10W to sputter (0.2 Å/s)
  - Final Cs evaporation, using SAES Getter Cs source

- Sample details
  - 1 MgO substrate (F009): 3 layers of 10 nm (QCM) thick each.
  - 1 Si substrate (F010): 1 layer of 30 nm thick (QCM)



Results – Sputter deposition

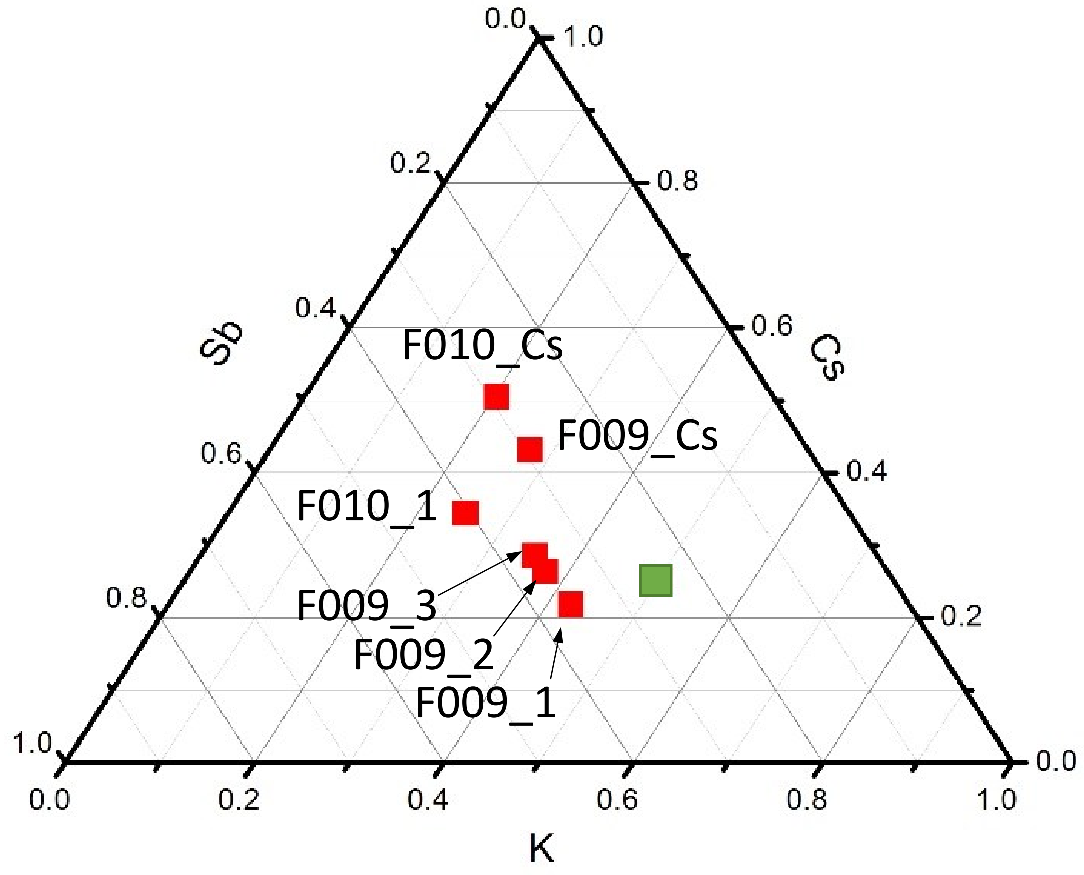
XRD images on F009 (MgO)

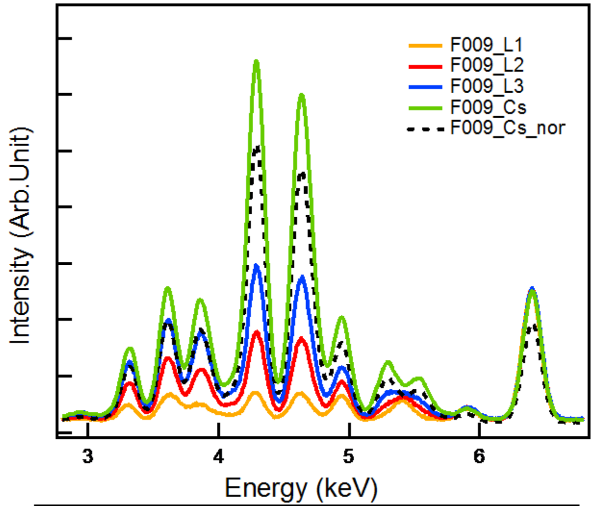


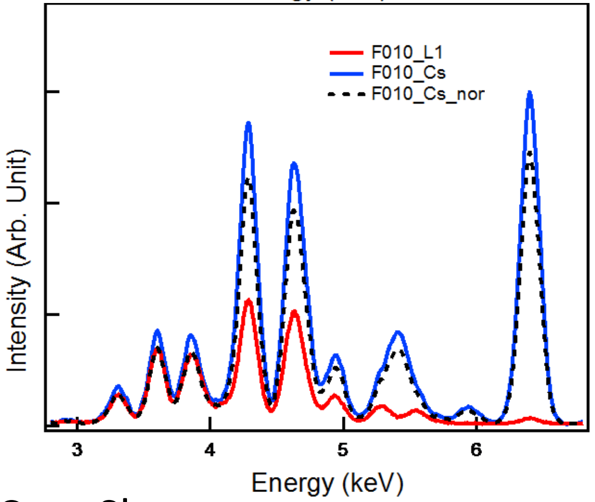
After sputter growth **After Cs deposition**

K_2CsSb rings identified with arrows

Results – Sputter deposition





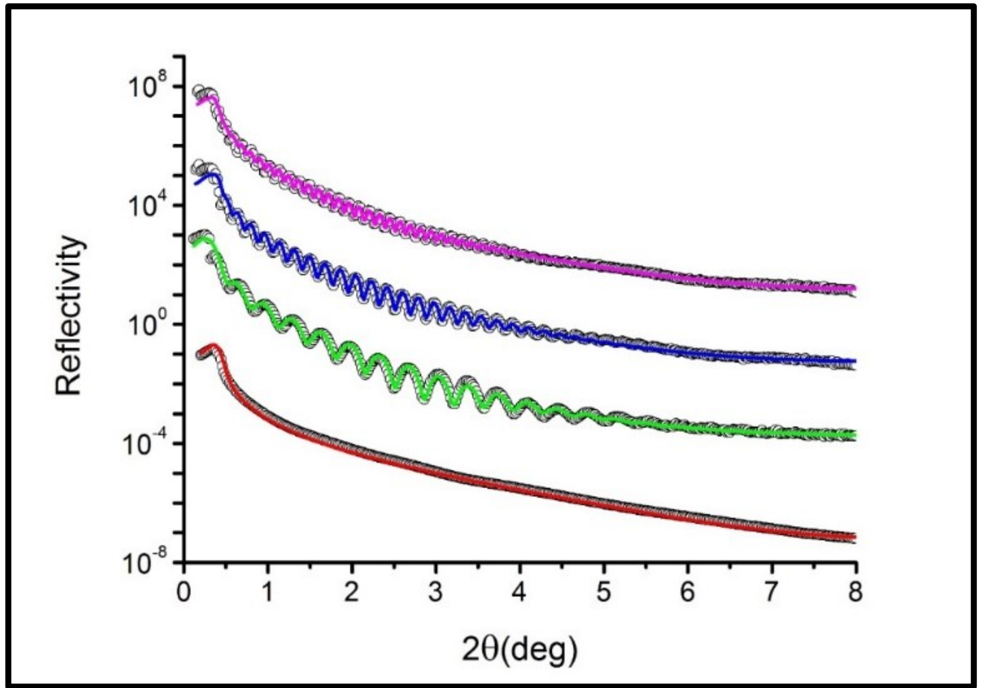


XRF result of sputtered sample.

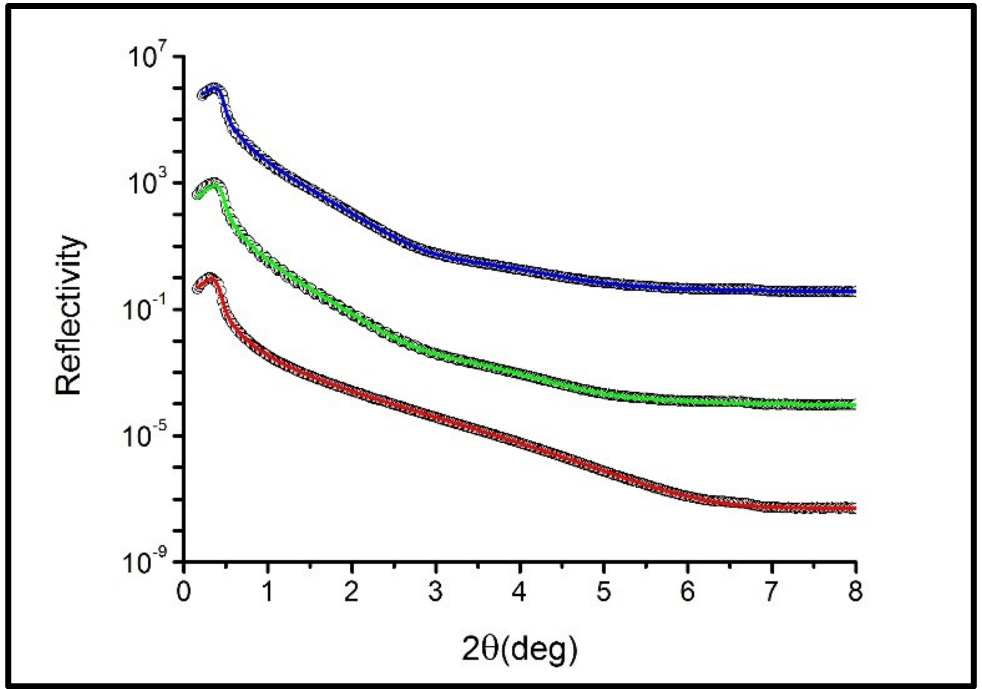
Chemical formula: F009: $K_{0.94}Cs_{1.47}Sb$; F010: $K_{0.7}Cs_{1.73}Sb$

QE @ 532 nm: 1%

Results



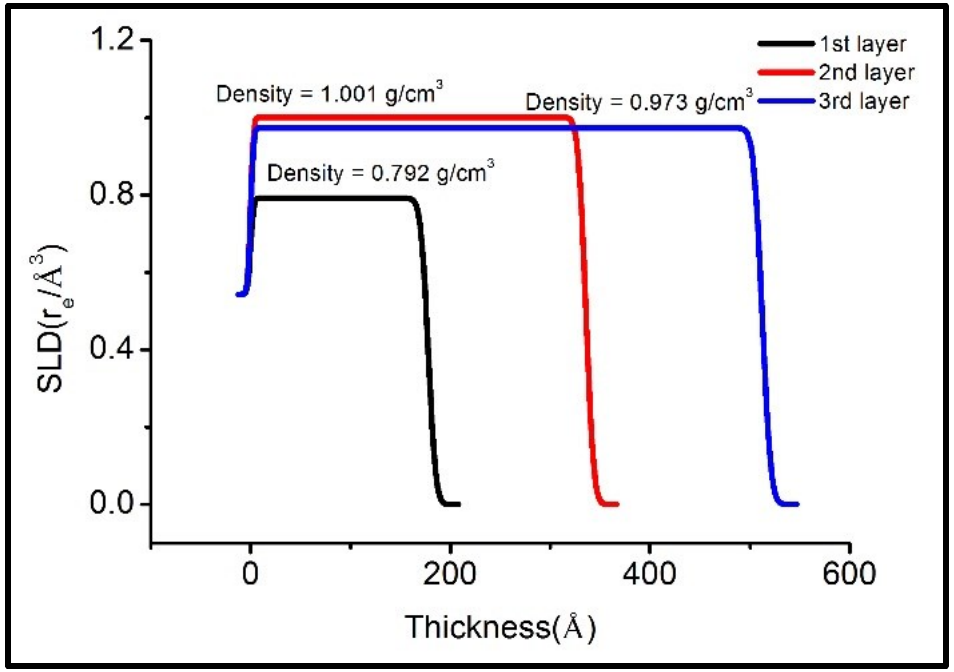
layer	Roughness(Å)	Thickness(Å)
3rd layer	6.87 (-0.17, 0.19)	512 (-1.17, 1.30)
2nd layer	6.20 (-0.19, 0.33)	336 (-1.35, 1.04)
1st layer	6.05 (-0.23, 0.18)	178 (-0.54, 0.47)
Substrate (MgO)	2.43 (-, -)	

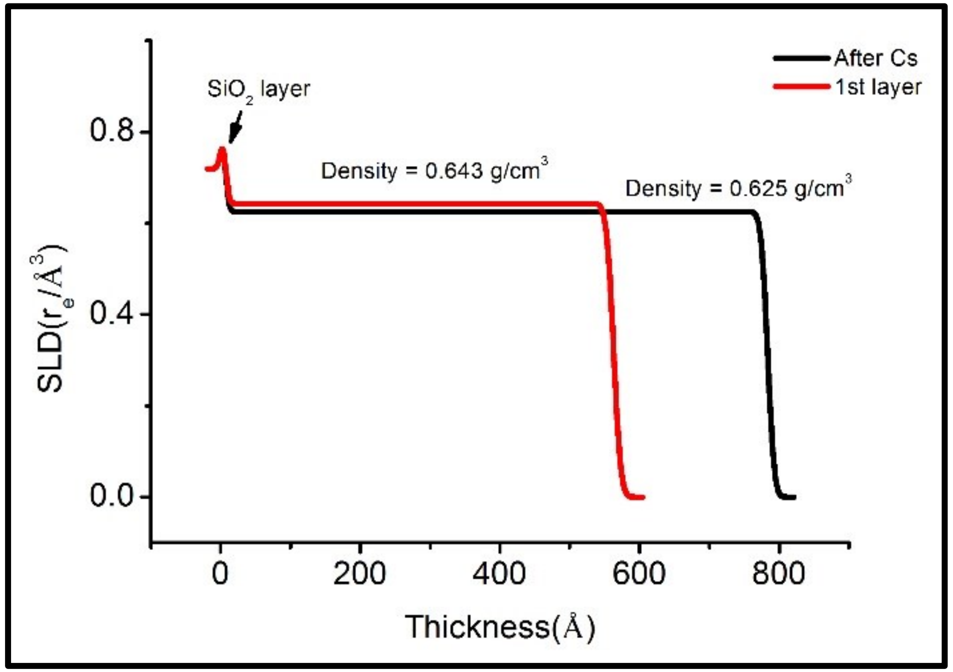


layer	Roughness(Å)	Thickness(Å)
After Cs	7.56 (-0.20, 0.13)	776.0 (-75.7, 71.0)
1st layer	8.21 (-0.068, 0.13)	555.4 (-2.6, 3.3)
Substrate (MgO)	3.93 (-0.03, 0.03)	

XRR simulation results of sample F009 (Left) and sample F010 (Right).

Results



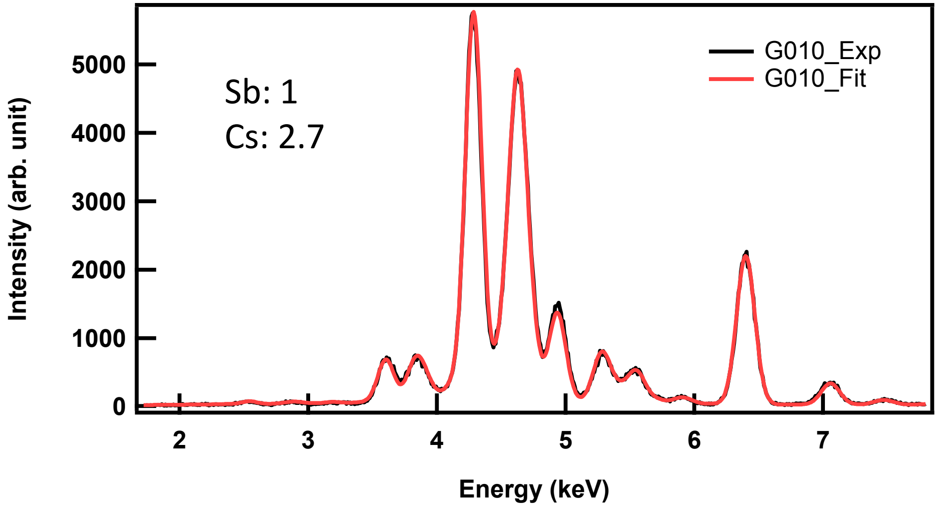


Simulated SLD (electron density) vs. thickness

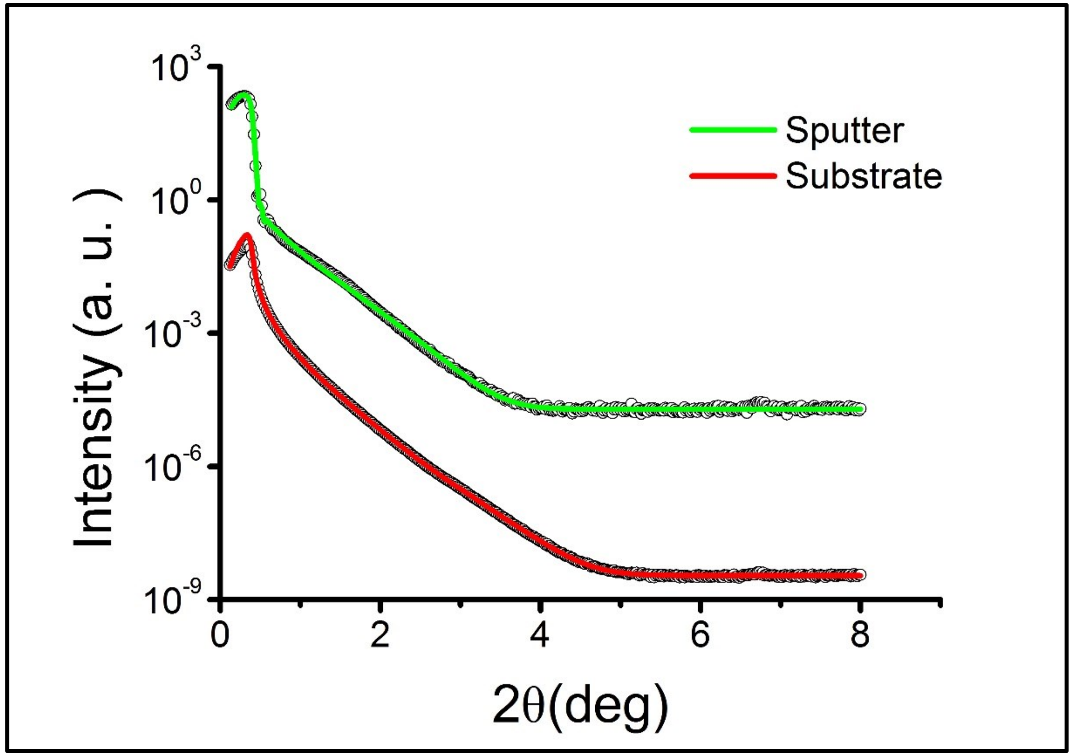
→ Two sputtered cathode exhibit higher electron density than evaporated cathode.

Results

Sputtered Cs-Sb @ 90℃



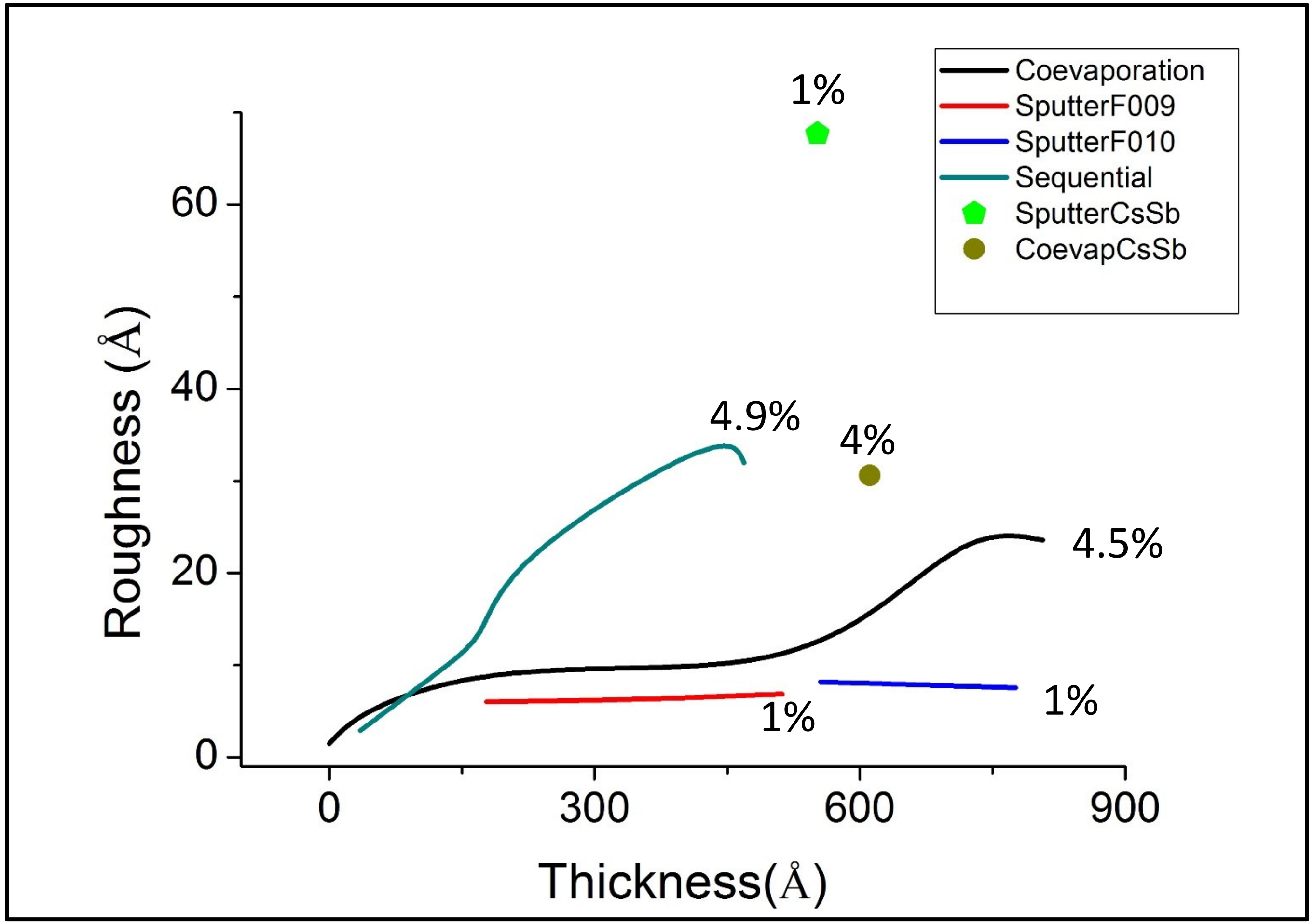
- XRF fitted result, chemical formula $Cs_{2.7}Sb$
- QE @ 532 nm: 1%



Layer	Thickness (Å)	Roughness (Å)
Sputtered Cs/Sb	552 (-21.1, +44.8)	67.7 (-3.6, +5.6)
Substrate	--	6.01 (-0.11, +0.14)

- XRR fitted result
- Uniform in electron density

Results



Roughness vs. thickness plot of alkali antimonide grown by different methods, with QE marked
Dots: Cs_3Sb; Lines: K_2CsSb

Summary

- Summary:
  - X-ray reflectivity is a useful in-situ technique in characterizing the roughness, thickness and density of photocathode growth, stoichiometry information obtained from XRF analysis helps the XRR simulation.
  - Co-evaporated CsK_2Sb/Cs_3Sb may end up in a smoother surface with the same quantum efficiency compared to sequentially-evaporated CsK_2Sb in previous study.
  - Sputtered CsK_2Sb exhibits the best surface roughness, with acceptable quantum efficiency. In comparison, sputtered Cs_3Sb have the same QE but is much rougher, further investigation needed.

- Acknowledgement:
  - Sincere thanks to my advisor Dr. John Smedley, to my colleagues: Mengjia Gaowei, Susanne Schubert, John Sinsheimer, Dr. Erik Muller, John Walsh, etc…
  - HZB colleagues: Martin Schmeisser, Julius Kuehn

Thank you for your attention!

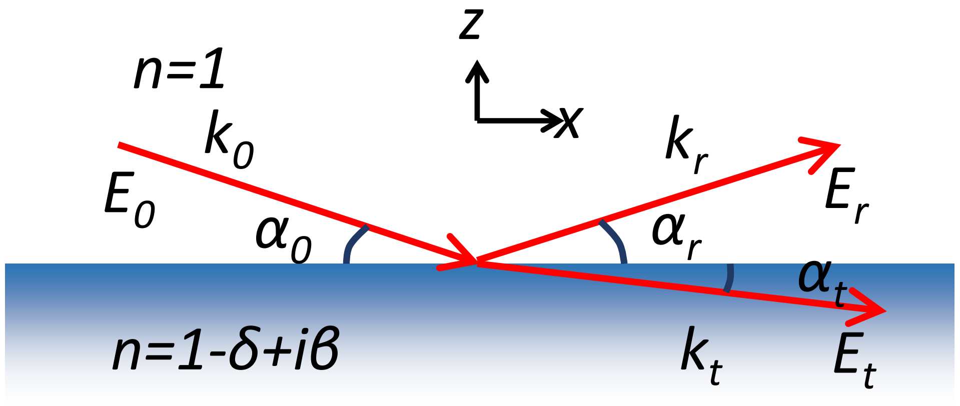
Basics of X-ray Reflectivity

- Interaction of X-ray with matter – Refractive index:

$$n=1-\delta+i\beta$$

Dispersive correction   Absorption correction

$$\delta=\frac{\lambda^2}{2\pi}r_e\rho_e \qquad \beta=\frac{\lambda}{4\pi}\mu$$



Critical angle for total reflection $\theta_c = \sqrt{2\delta}$

- Fresnel reflectivity
- <u>Amplitude</u>

$$r(\theta) = \frac{\theta-\sqrt{\theta^2-\theta_c^2}}{\theta+\sqrt{\theta^2-\theta_c^2}}$$

<u>Intensity</u>

$$\mathrm{R}(\theta) = \left|\frac{\theta-\sqrt{\theta^2-\theta_c^2}}{\theta+\sqrt{\theta^2-\theta_c^2}}\right|^2$$

- Plus absorption of the X-ray beam

Fresnel reflectivity becomes:

$$\mathrm{R}(\theta) = \left|\frac{\theta-\sqrt{\theta^2-\theta_c^2-2i\beta}}{\theta+\sqrt{\theta^2-\theta_c^2-2i\beta}}\right|^2$$

→ Reflectivity curve can be calculated based on the electron density and the absorption coefficient of the material.