Development of Multialkali antimonides photocathodes for high brightness photoinjectors

Sandeep Mohanty on behalf of PITZ & LASA team, European Workshop on Photocathodes for Particle Accelerator Applications (EWPAA 2022), Sep 20 – 22, 2022,Milan

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- Summary & Future plan

S.Mohanty, H. Qian,M. Krasilnikov, A. Oppelt, F. Stephan

DESY, Zeuthen, Germany

D. Sertore, L. Monaco, G. Guerini Rocco

INFN LASA, Segrate, Italy and Università degli Studi di Milano

INFN LASA
Istituto Nazionale di Fisica Nucleare
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DESY.

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Introduction to the growth procedures

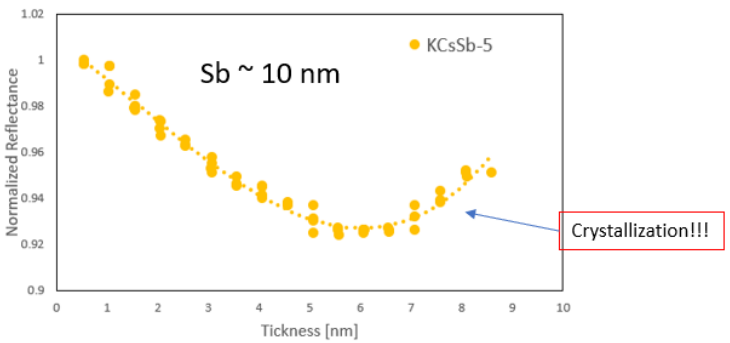
KCsSb sequential growth analysis

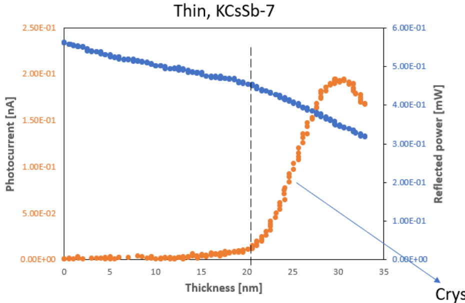
Various experimental results show formation of islands of Sb on Mo surface and coalesce to a monolayer and **crystalize** at around **4 - 8 nm** at around.

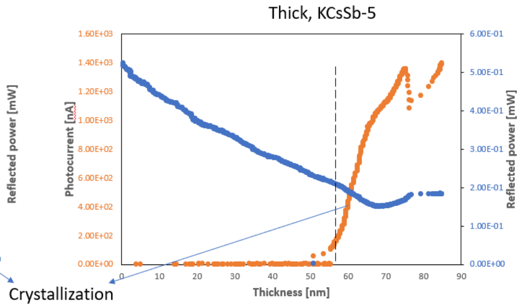
ref: C.Ghosh Journal of Applied Physics 49, 4549 (1978)

Sb ➕

We put 5 or 10 nm







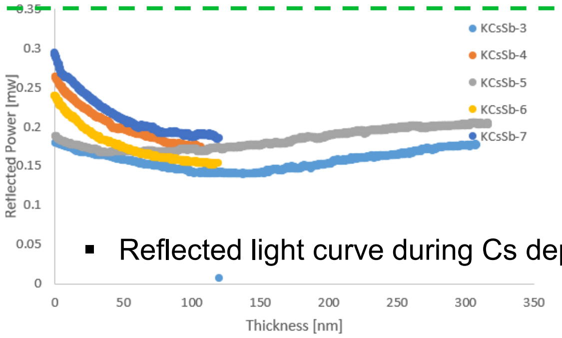
K → K_3Sb ➕

Untill reach the Photocurrent yield

Phase 1

Formation of K3Sb with cubic and hexagonal orientation

Ref: S.g Schubert Conference: 5th International Particle Accelerator Conference IPAC14

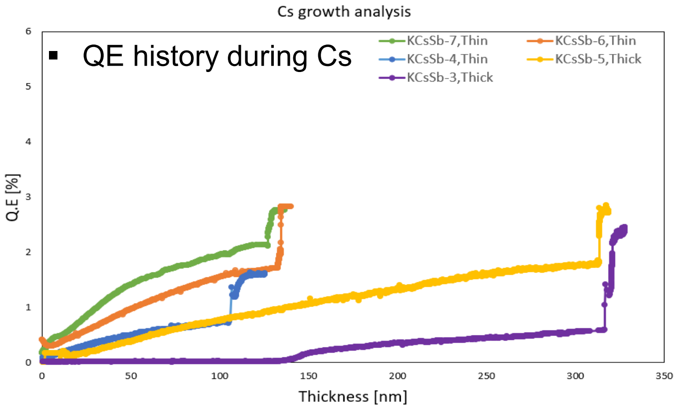


Cs → KSbCs

Untill reach the Photocurrent yield

Phase 2

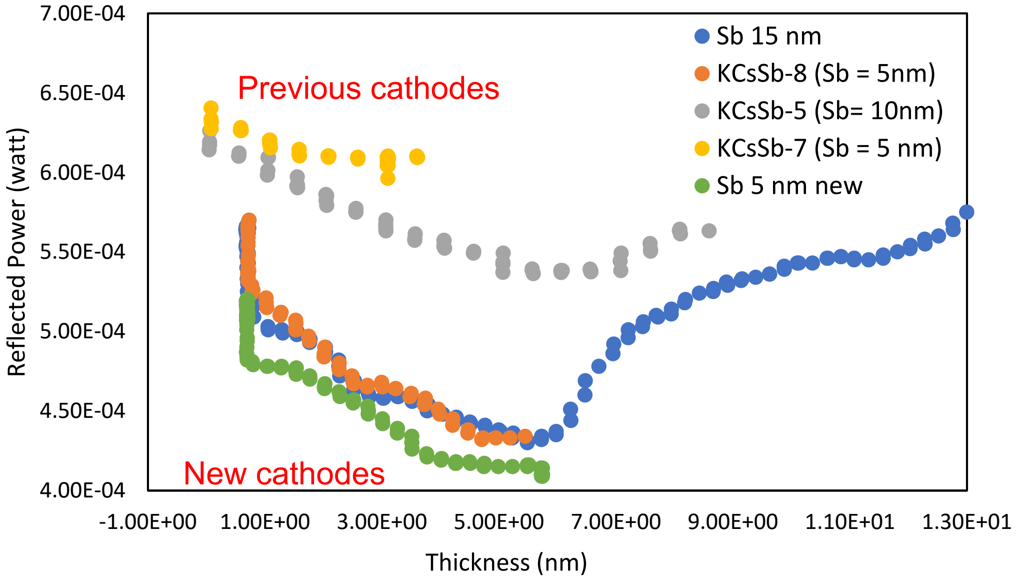
Formation of KSbCs



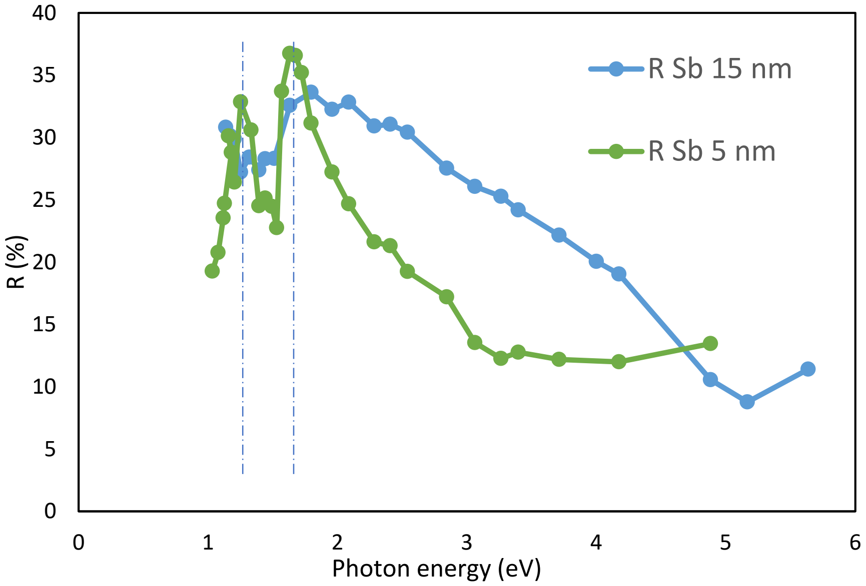
Sb 5nm = Thin
Sb 10 nm = Thick

Sb phase (crystalline and amorphous)

- We usually deposited initially **Sb of 5 or 10 nm** on Mo substrate.
- The reflected power curve turns upward at around 6-7 nm (also depends upon various other factors like sub. temp., surface quality, etc.), showing the transition from amorphous to crystallization.
- The behavior of the Reflectivity curve of both the compound (Sb = 5 & 10 nm) appeared differently.



- Real-time reflected power curve during Sb deposition



- Reflectivity

* Here the transmittance value of viewport is not included in QE and reflectivity data

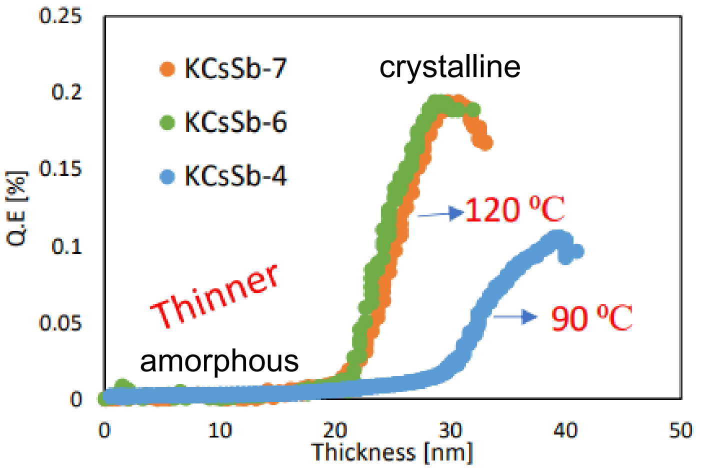
- Previous XRD studies showed also a similar behavior of Sb, i.e., a transition that happened from amorphous and crystallization in Sb happened between 4-7 nm [1].

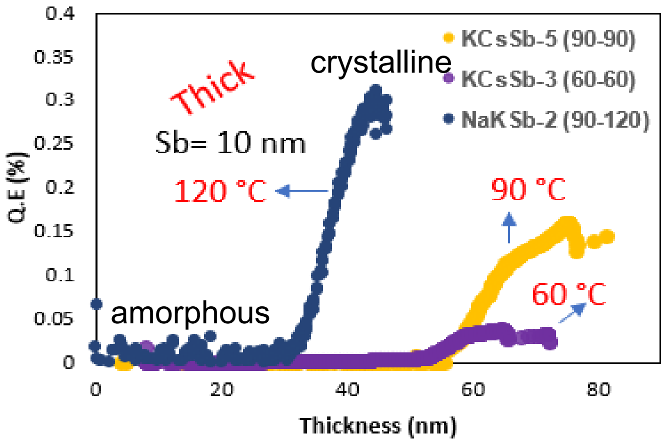
1. Ruiz-Oses M, Schubert S, Attenkofer K, Ben-Zvi I, Liang X, Muller E, Padmore H, Rao T, Vecchione T, Wong J, Xie J and Smedley J 2014 APL Mater. 2 121101

KSb phase

- Various factors affecting the film properties: substrate temperature, deposition rate, surface quality, etc.

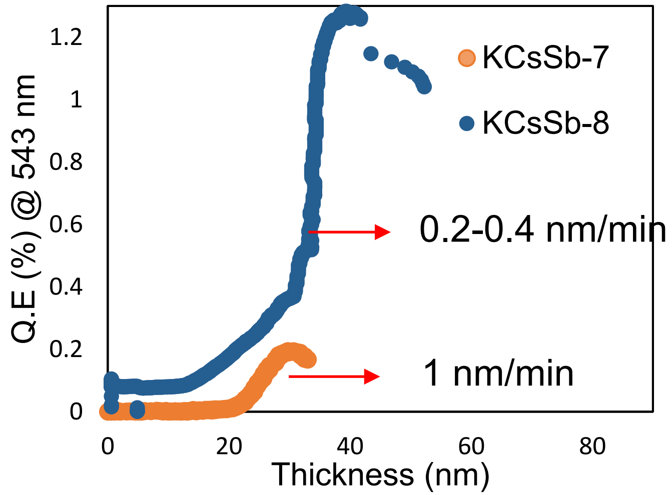
Substrate temperature dependent



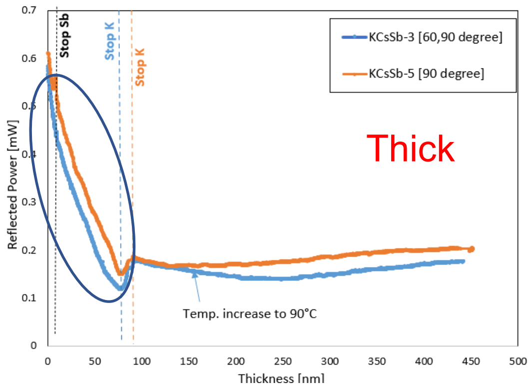


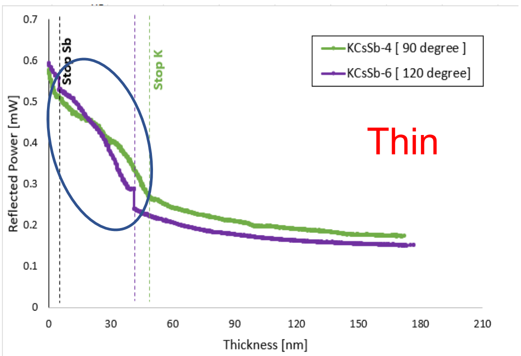
- Faster diffusion process between Sb and K due to a higher temperature, induces a faster crystal growth rate in the KSb film.

Deposition rate



✓ The maximum value of **1.5%** @ 515 nm QE is achieved for the KSb compound



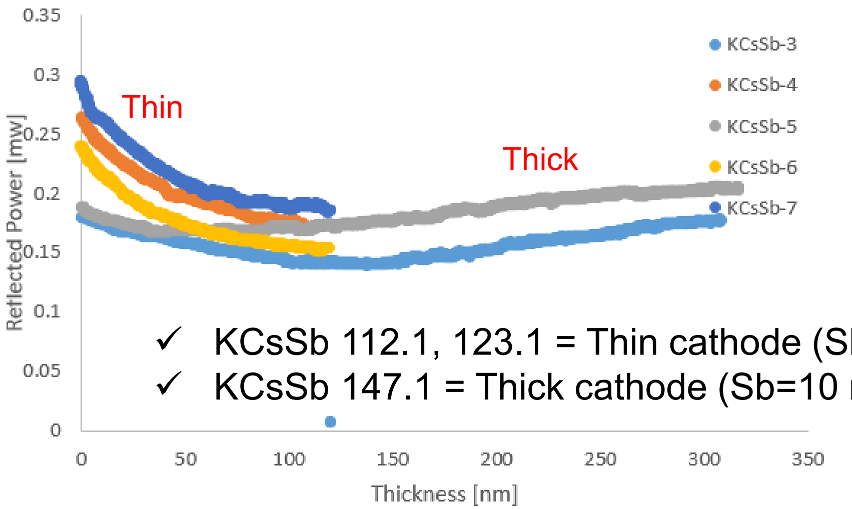


- Real-time reflected power curve during deposition

✓ KCsSb 112.1, 123.1 = Thin cathode (Sb= 5 nm)
✓ KCsSb 147.1 = Thick cathode (Sb=10 nm)

KSbCs phase

- Difference in reflected power curve between thin & thick cathodes shows the potential formation of different crystal orientations in each case (needs to verify with surface characterization study!).



✓ KCsSb 112.1, 123.1 = Thin cathode (Sb= 5 nm)
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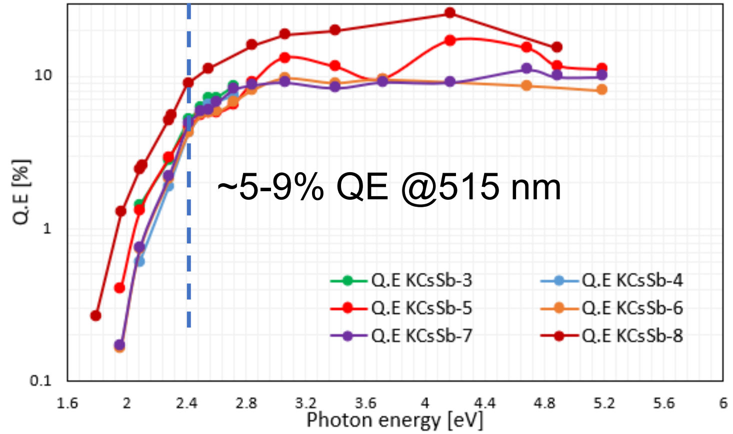
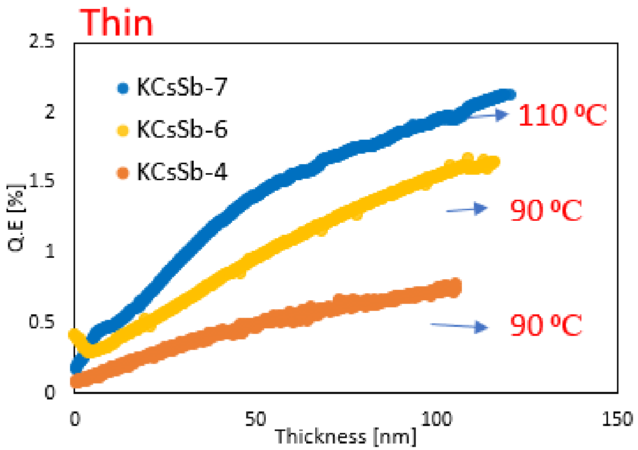


- Spectral response

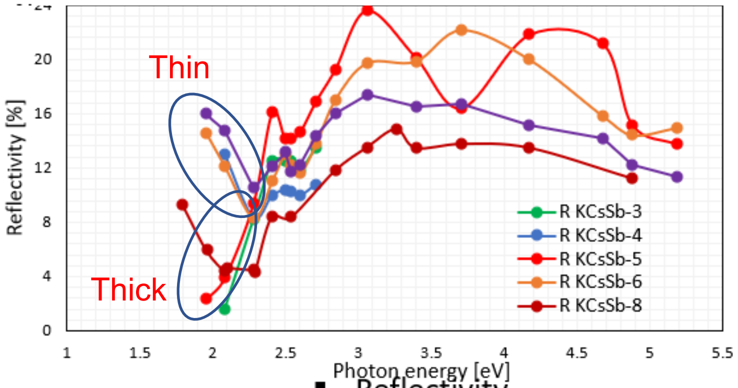
- Real-time reflected power curve during Cs deposition



Substrate temperature dependent

KCsSb-3 (Thick)

KCsSb-4 (Thin)



- Reflectivity

* Here the transmittance value of viewport is not included in QE and reflectivity data

➢ 8 cathodes (prototype) produced so far with sequential deposition in the **R&D process**, max Q.E @514 nm is recorded ~ 9 % with peak Q.E around 25% @ 297 nm.

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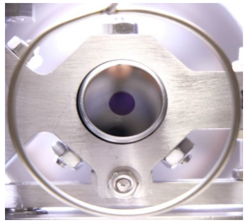
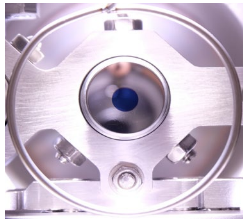
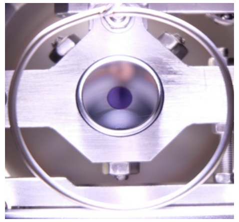
Cathode Production and Test in the RF Gun at PITZ

Cathode preparation

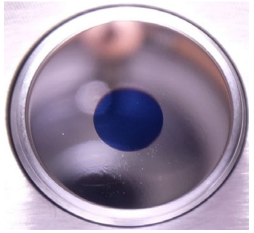
- Successfully produced the first batch of green cathode in “new production system” at INFN LASA.
  - ✓ Total **3 cathodes** produced with **sequential deposition**.
  - ✓ Out of which **1 thick** (147.1) (Sb= 10 nm) and **2 thin** (112.1, 123.1) (Sb = 5 nm) cathodes.
  - ✓ **Q.E @514 nm** is recorded **4-8 %** for thick and thin cathodes respectively after the production.
  - ✓ All the cathodes has survived during the cathode box transportation, installation and cathode insertion.
  - Estimated the **Eg+Ea** value of grown cathode is around **1.8 eV,** slightly lower than the literature value (1.9 eV, 2.1 eV).
  - Behaviour of reflectivity was different for thin and thick cathode at higher wavelength (low energy) (similar like R & D experience).


112.1


147.1


123.1

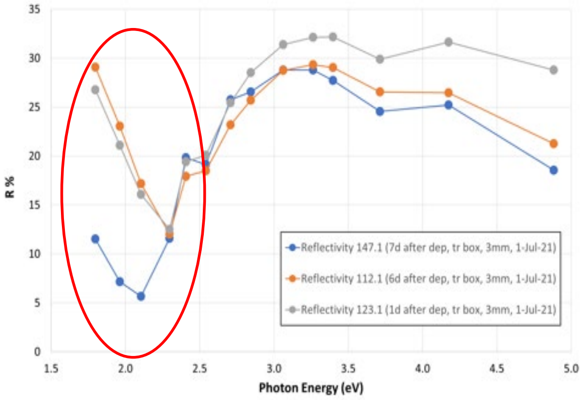

Violet, thin


Blue, thick

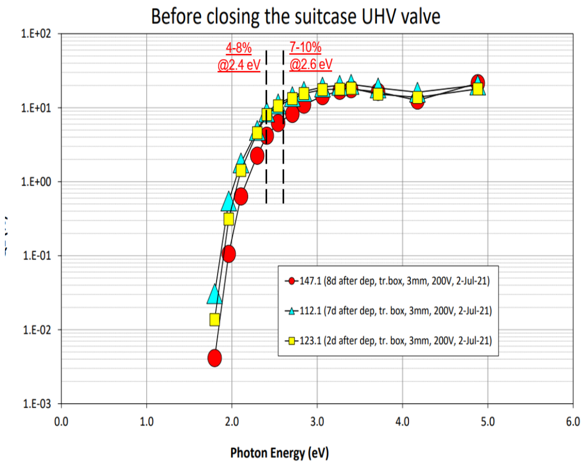

Violet, thin

Cathode	Sb evaporation*	K evaporation*	Cs evaporation*	K/Sb ratio	Cs/Sb ratio	QE @ 543nn (after producti
147.1	120 °C, 4nm	150 °C, 31.4nm	135 °C, 89.1nm	7.85	22.27	3.9%
112.1	120 °C, 2.9nm	150 °C, 22.3nm	140 °C, 76.8nm	7.69	26.48	3.3%
123.1	120 °C, 2.9nm	150 °C, 22.4nm	150 °C -> 120 °C, 78.3nm	7.72	27	4.1%

*All the thickness values are read with a quartz microbalance positioned out of the deposition axis.



- Reflectivity

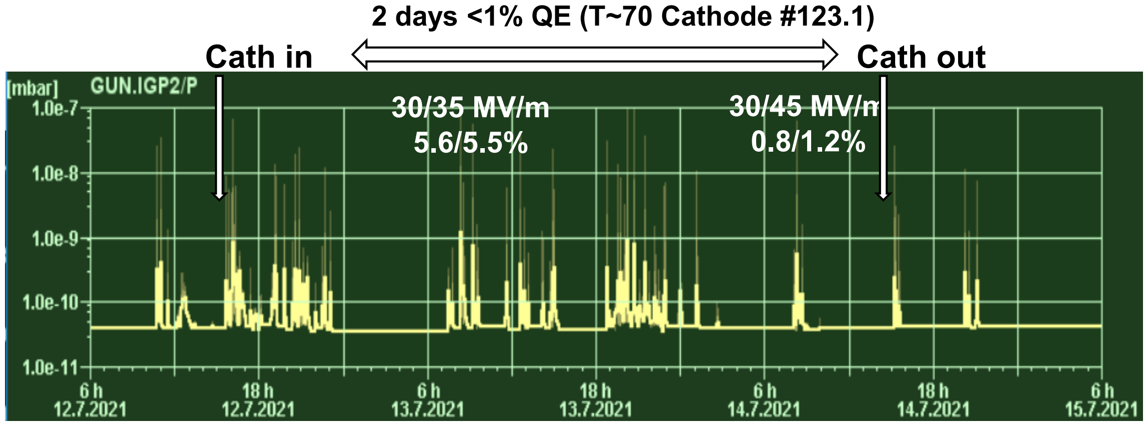


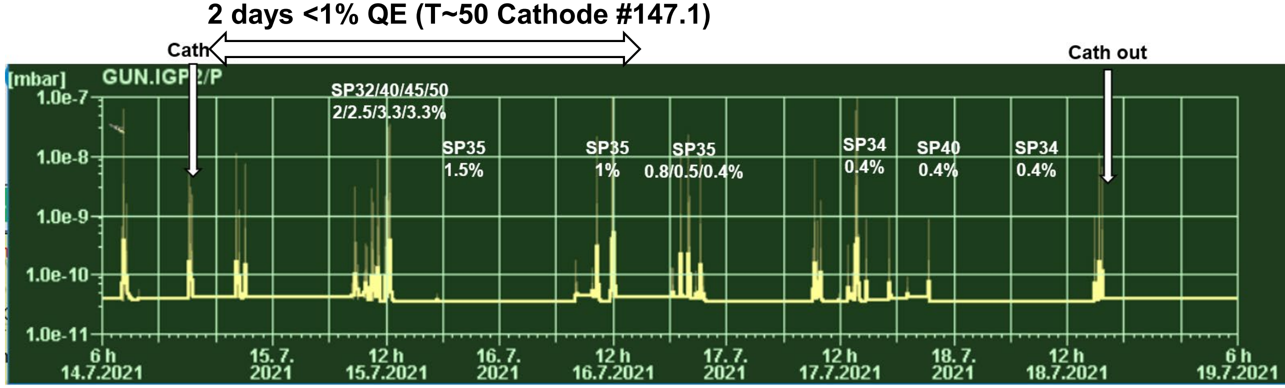
- Spectral Response

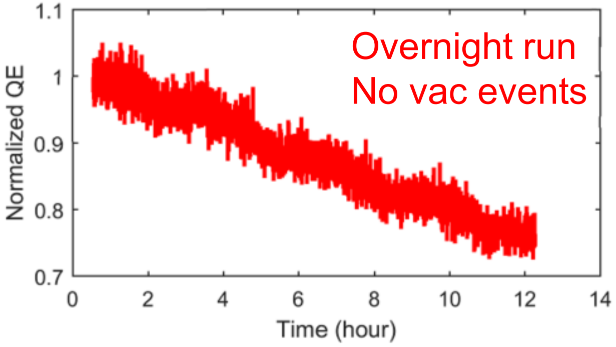
Cathode under high gradients

Cathode RF conditioning, QE, life time, vacuum

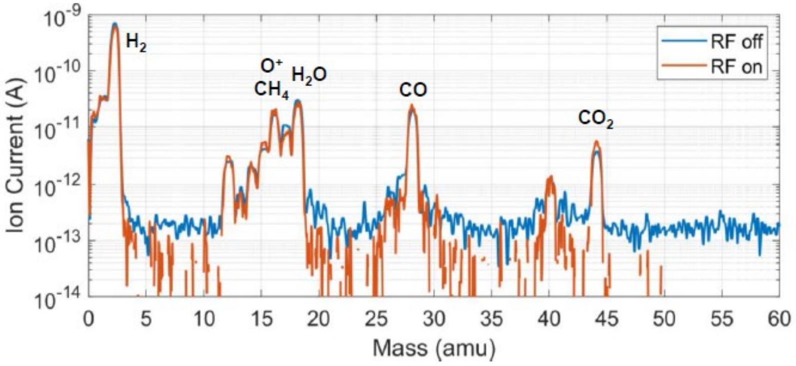
- Cathode RF conditioning
  - Below 30 MV/m, almost not necessary, up to 400 us was tested without vac events
  - Above 30-40 MV/m, much more vac events than Cs_2Te conditioning, degrades QE significantly
- Cathode QE
  - Fresh QE in gun is consistent with LASA measurements
  - QE decrease dominated by vac events during conditioning, QE also slowly decreases w/o vac events.



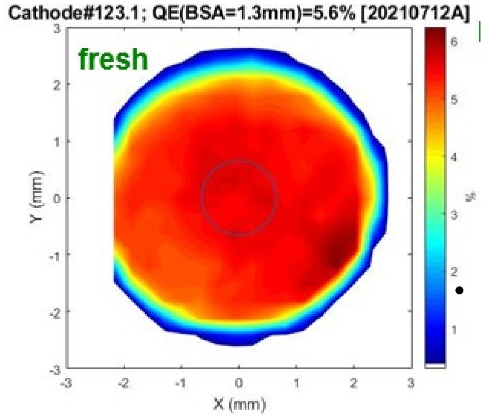




QE slowly drops by 20% in 12 hours



2 days <1% QE (T~50 Cathode #112.1)



- Uniformity in QE

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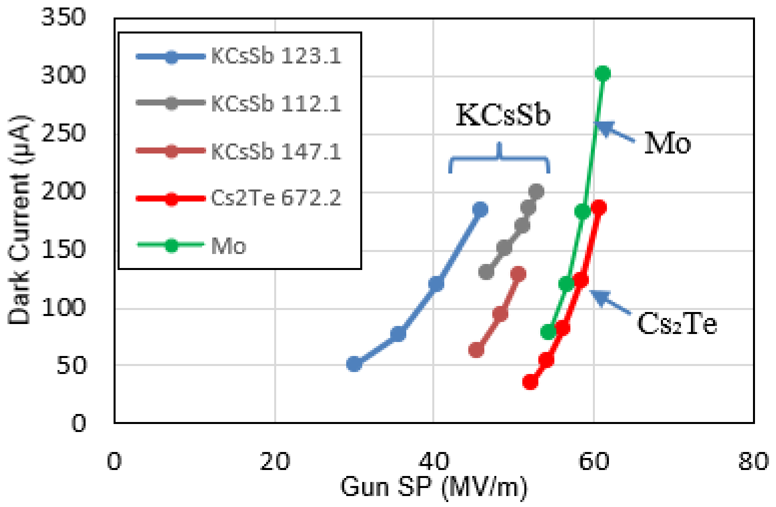
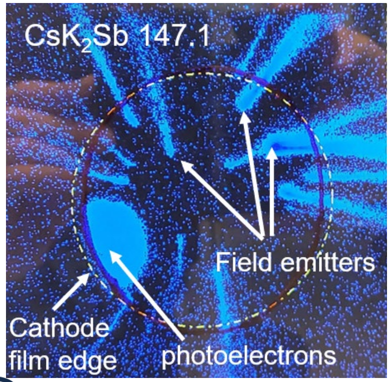
Cathode under high gradients

- Dark current vs gun gradient



- All three cathodes have a higher dark current compared to Cs2Te cathodes, Major part of the contribution is due to the lower work function of **CsKSb = 2.1 eV** compared to **Cs_2Te = 3.5 eV**.

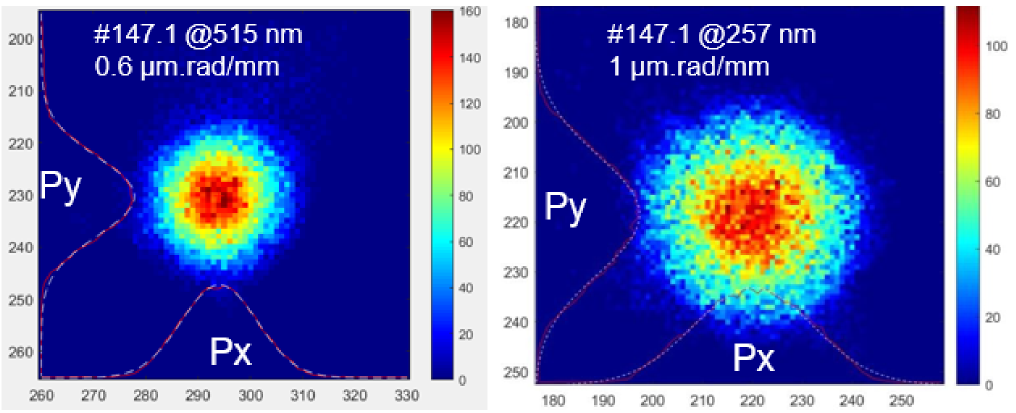


Dark current imaging

	beta	A (nm²)
Cs_2Te 672.2	126	15212
123.1	462	0.002
147.1	97	0.98
147.1	102	1.1
112.1	99	0.3
112.1	246	0.013

- F-N analysis, high dark current due to low emission threshold, not due to surface quality

- Thermal emittance vs QE and laser wavelength
  - 515 nm, 19 MV/m, **0.6 µm.rad/mm** (2% & 0.8%, 1.5%)
    - Consistent with APEX gun results
  - 515 nm, 29 MV/m, 0.7 µm.rad/mm (1.5%)
    - >30 MV/m not possible due to high dark current
  - 257 nm, 19 MV/m, **1 µm.rad/mm** (2.5%)
    - Very similar to Cs_2Te cathode

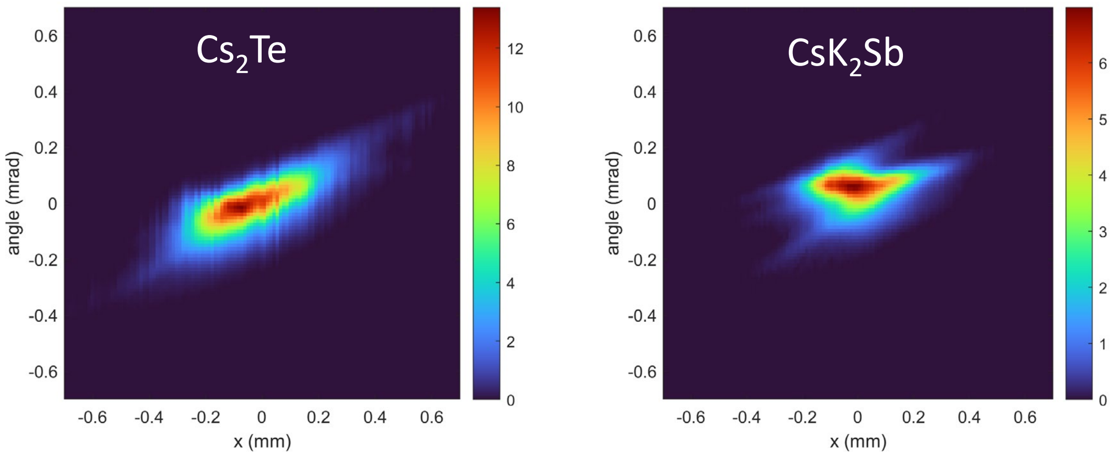


- 2D distribution of photoemission transverse momentum

100 pC emittance and response time

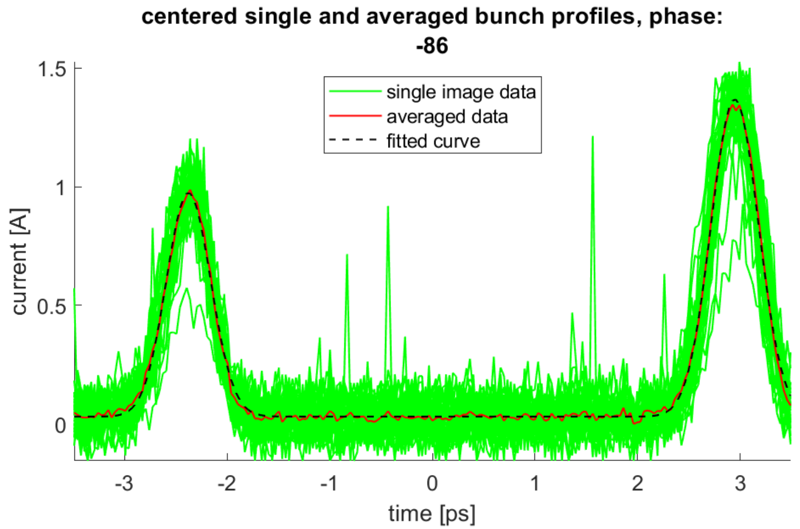
- **100 pC** emittance, CsK_2Sb vs Cs_2Te
  - **40 MV/m** 1.6 cell L-band gun, ~20 MeV linac
  - Emittance reduced by **~23%**, 4D brightness increased by **~60%**

Measurements	**Cs_2Te 1 µm.rad/mm**	**CsK_2Sb 0.7 µm.rad/mm**	Unit
95% rms emit.	0.36	0.28	µm.rad
Gauss emit.	0.33	0.25	µm.rad
4d brightness	760	1209	$pC/(\mu m.rad)^2$



- **100 pC beam phase space**

- Cathode response time measurement
  - #147.1 CsKSb, **measured when QE dropped to 0.4%**
  - Two lasers with known optical delays shine the cathode to calibrate the beam temporal response at the cathode.
  - Preliminary data shows response time is **below the resolution of 100 fs!** Much shorter than high QE Cs2Te cathode response time in UV (~200 fs)



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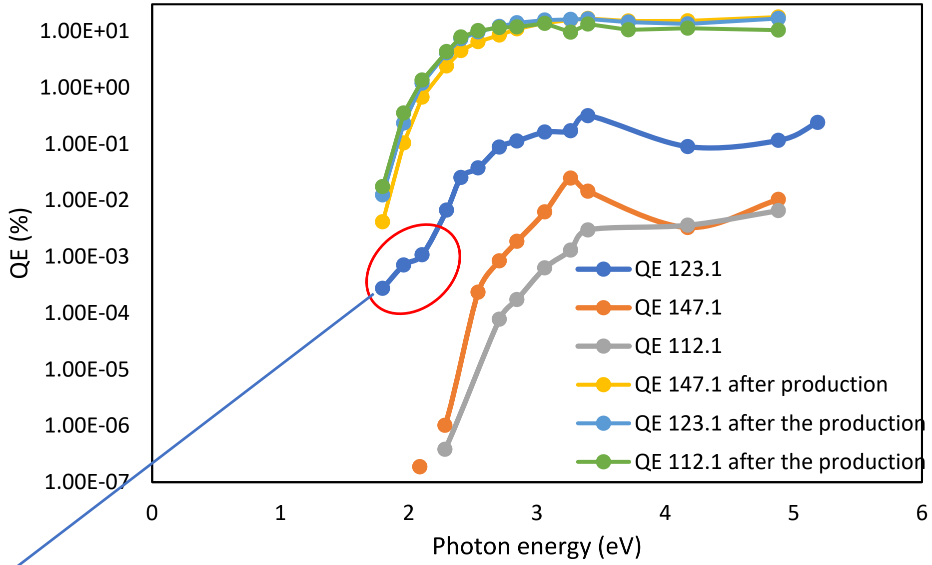
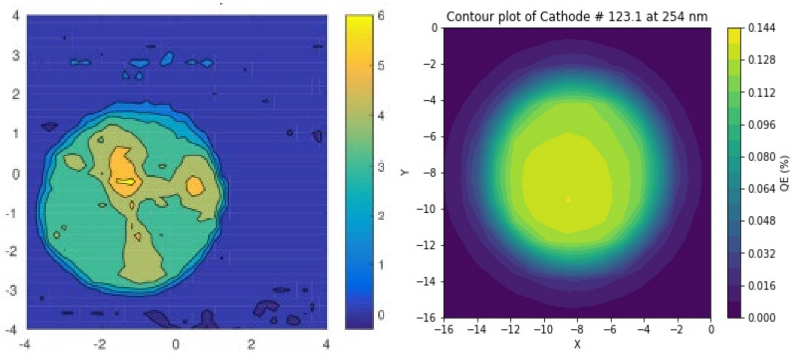
Post-operation optical diagnostic of "tested" KCsSb cathodes

- Spectral response + reflectivity measurements have been done for all three cathodes.
- Photoemission threshold (Eg+Ea) increased from 1.8 eV to **2.08 eV** for cathode #147.1 and #112.1 (**similar like Cs_2Te**), however, in the #123.1 case, it is different.
- The spectral response behavior of "used" photocathodes shows the potential **oxidation** of cathode films.
  - ✓ KCsSb 112.1, 123.1 = Thin cathode (Sb= 5 nm)
  - ✓ KCsSb 147.1 = Thick cathode (Sb=10 nm)



- Spectral response

515 nm 254 nm



- QE maps of Cathode #123.1 after the usage

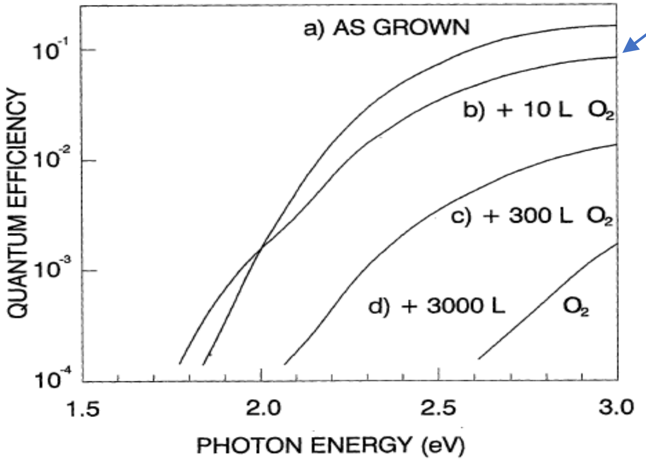
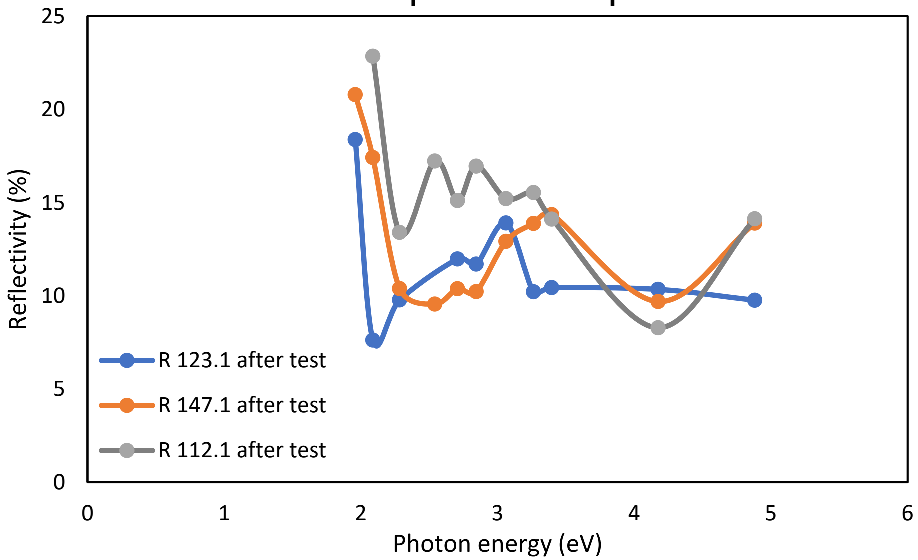


Fig. 7. Spectral response curves of photocathode $Cs_{2.5}K_{0.5}Sb$ (a) as grown; (b) after 10 L oxygen exposure; (c) after 300 L oxygen exposure; (d) after 3000 L oxygen exposure. The quantum efficiency is given per incident photon.



Leonardo Soriano and Luis Galán Luis Galán 1993 *Jpn. J. Appl. Phys.* **32** 4737

- Reflectivity

Operational and Post-usage Summary

- The results in terms of QE, thermal emittance, and response time are very promising. However, the limiting factors are high dark current and short operational lifetime.
- Post-usage analysis shows different levels of oxidation of the cathode films.

➢ Future Plans:

- To overcome these drawbacks, we are currently improving our cathode recipe. A multi-wavelength technique will be added for real-time monitoring of QE and reflectivity during cathode formation.
- The new co-evaporation technique will be introduced in our production system to explore its effect on the operation of our photocathodes in RF guns in the future.

New Optical characterization technique (Refracting index)

- Fresnel equation was used to calculate the index of refraction of Molybdenum plug (#137.1)

$$R_p = \left| \frac{n_1 \cos\theta_t - n_2 \cos\theta_i}{n_1 \cos\theta_t + n_2 \cos\theta_i} \right|^2 = \left| \frac{n_1 \sqrt{1 - \left(\frac{n_1}{n_2}\sin\theta_i\right)^2} - n_2 \cos\theta_i}{n_1 \sqrt{1 - \left(\frac{n_1}{n_2}\sin\theta_i\right)^2} + n_2 \cos\theta_i} \right|^2.$$

n_1= refracting index of vacuum =1 , n_2= refracting index of substrate, θ= angle of incidence , Rp = reflectivity of p-polarized light

- Refracting index (n) of KCsSb #8 :
  - **1.18** @ 543 nm (compare to reference 2 and 3.6 @ 533 nm)
  - **1.4** @ 514 nm

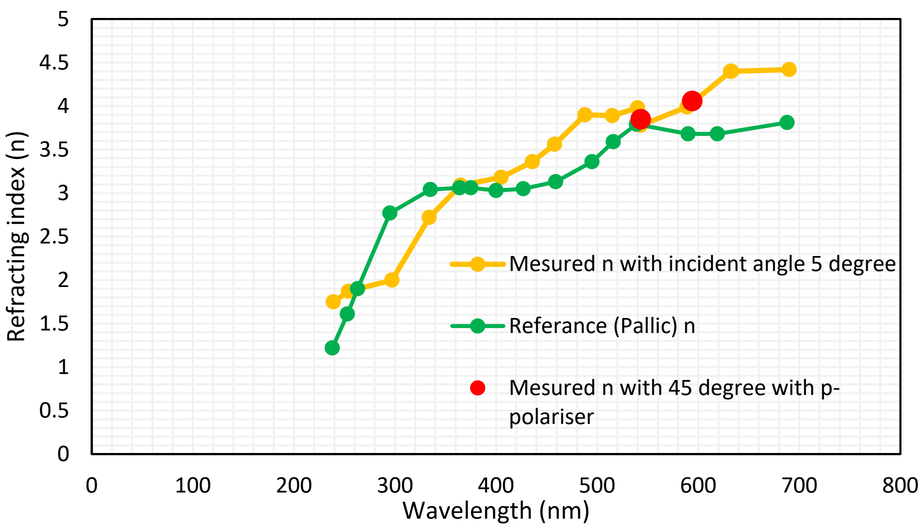
*Assume specular reflection from a smooth photocathode surface, and ignore the possibility of enhanced reflectivity due **to multiple reflection**s within the photocathode thin film

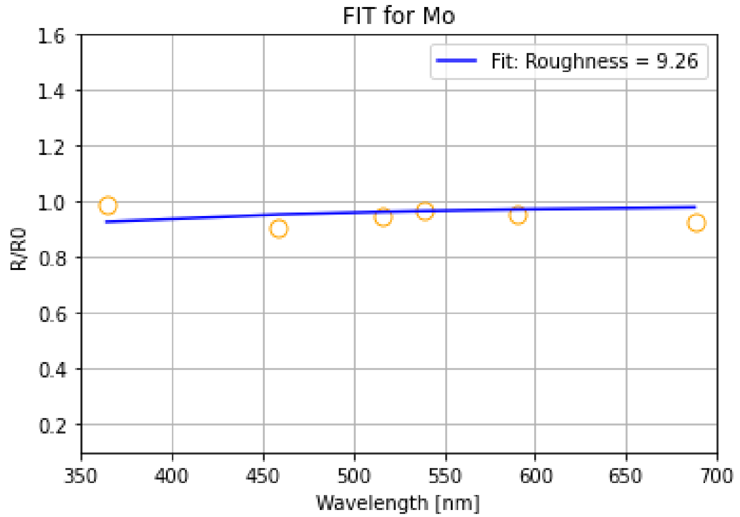
$$\frac{R}{R_0} = \exp\left(-\left(\frac{4 \cdot \pi \cdot \cos(\theta) \cdot \sigma}{\lambda}\right)^2\right) \quad [1]$$

R_0= reference reflectivity, θ=angle of incidence
R = measured reflectivity , σ= average roughness

- Avg. roughness of Mo plug #137.1 = **9 nm**

- Refracting index





- Roughness estimation

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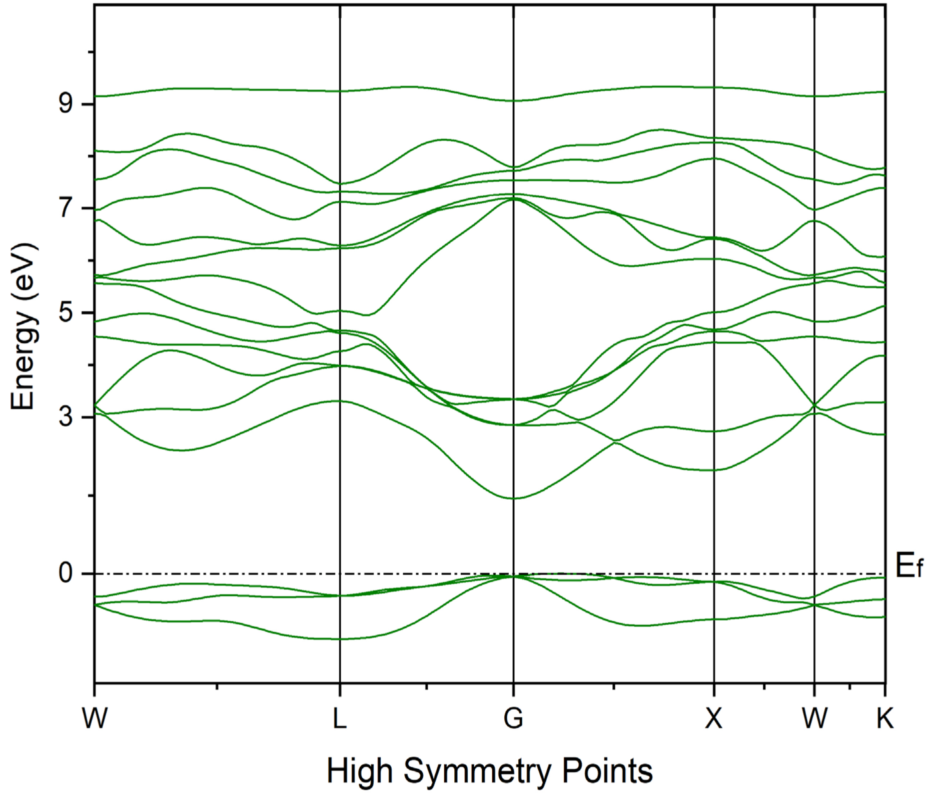
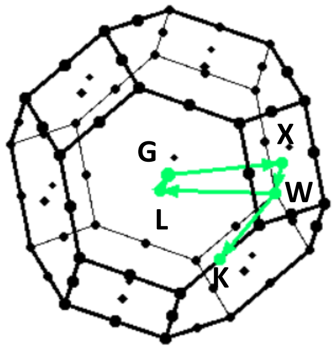
K_2CsSb electronic structure (DFT study)



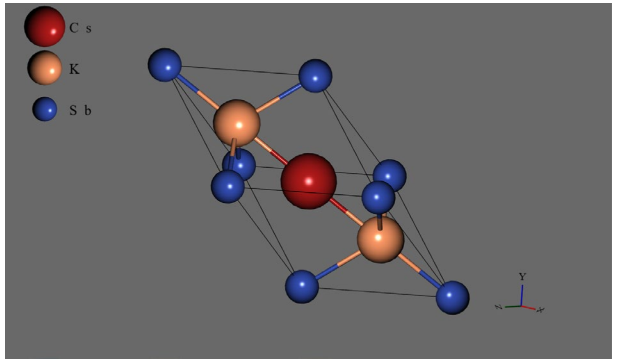
- Band structure calculation by HSE method

	DFT (G-G) direct gap	HSE (G-G) direct gap
Egap (K points)	0.8446 eV	1.49 eV

Experimental band gap = **1.2 eV**

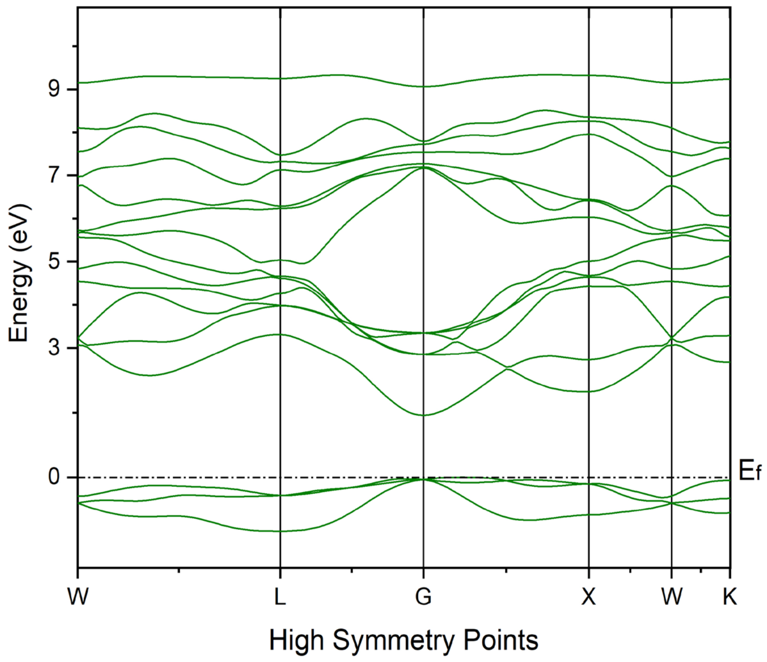


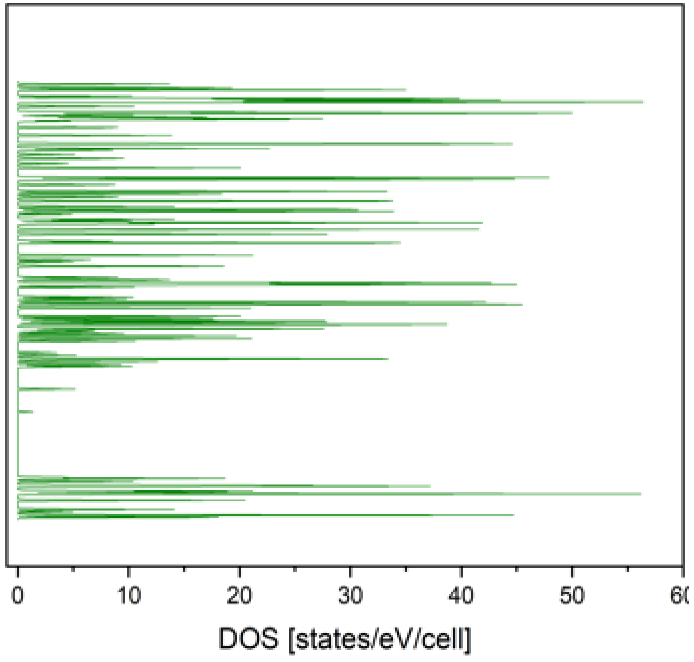
a = 8.7587 angstrom

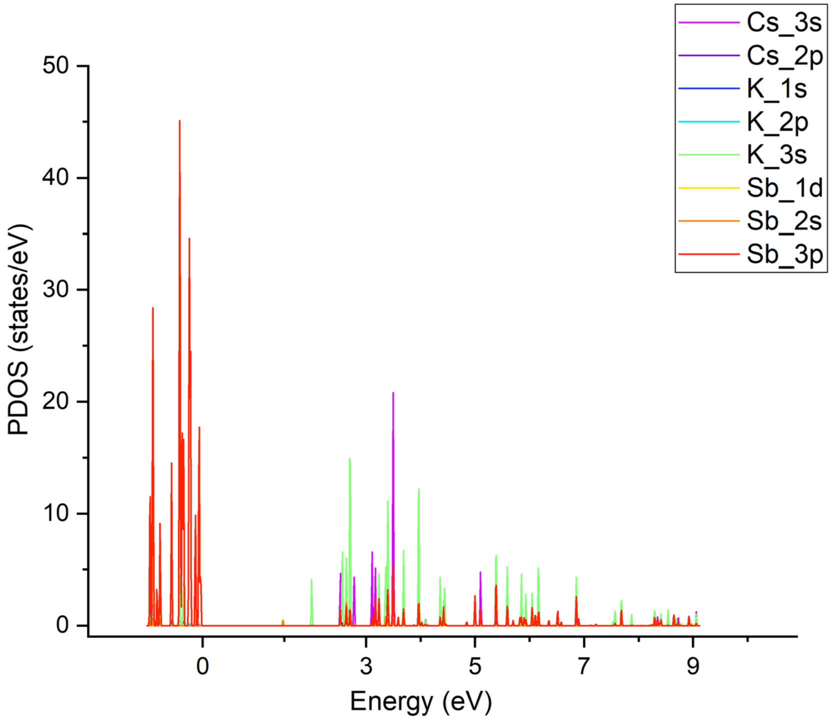


K_2CsSb electronic structure (DFT study)

- Ground states, DOS, and PDOS are calculated by QUANTUM ESPRESSO (HSE hybrid function) and the band structure is analyzed by using Wannier90 tool.







- PDOS calculation reveals that the valence bands are dominated by the partially filled Sb-p state.
- While, in the conduction band it carries the imprint of each compound.

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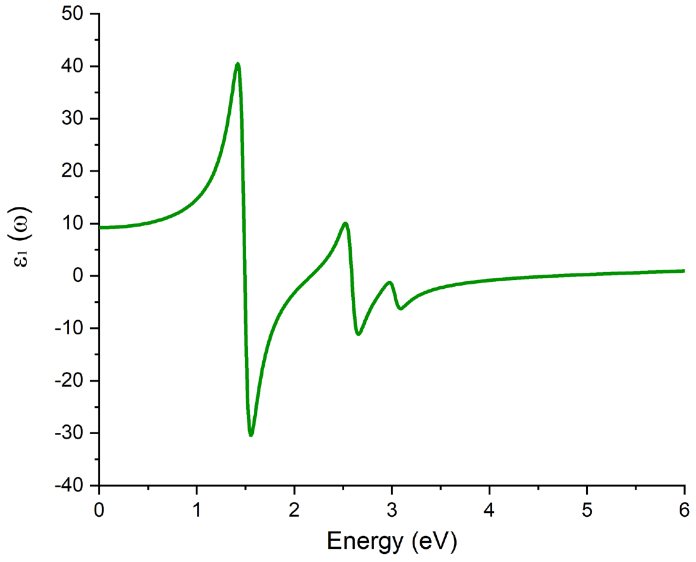
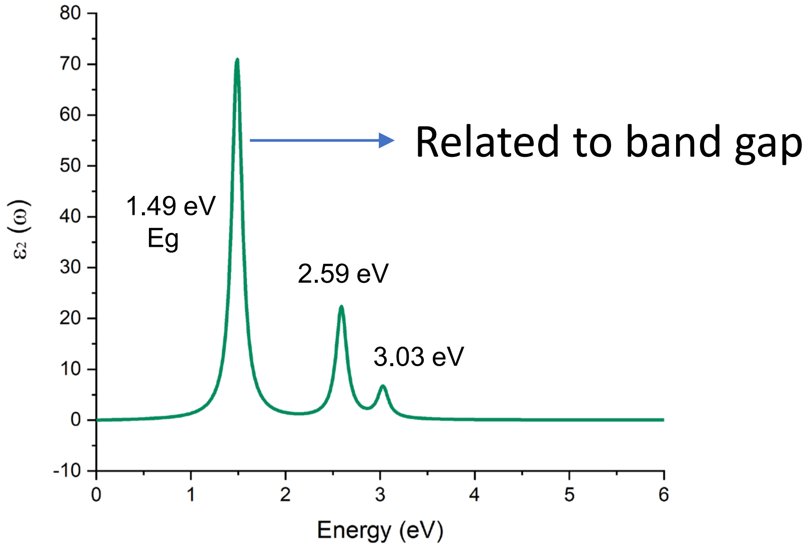
K_2CsSb Optical Properties(DFT study)

- Ground-state calculations, are implemented in Quantum Espresso and the dielectric function is calculated in epsilon.x post-processing utility.

- Real part of the dielectric function



- Imaginary part of the dielectric function



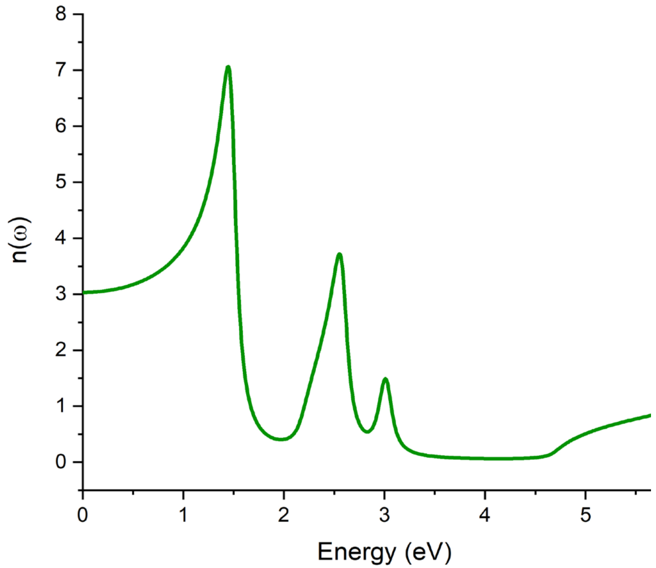
- Intra-band transitions i.e. either conduction-to-conduction or valence-to-valence transitions

- Inter-band transitions i.e. transitions from VBs to CBs

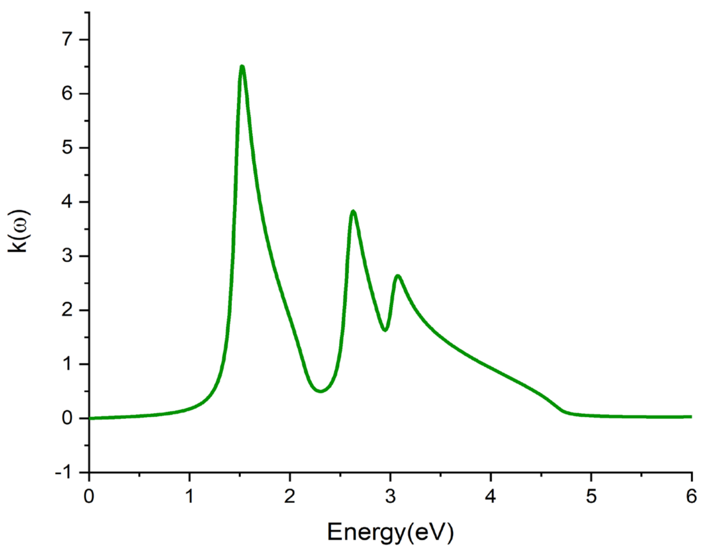
K_2CsSb Optical Properties(DFT study)

- $n(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}-\varepsilon_1(\omega)}{2}\right]^{0.5}$
- $k(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}+\varepsilon_1(\omega)}{2}\right]^{0.5}$
- $R(\omega) = \frac{(1-n)^2+k^2}{(1+n)^2+k^2}$

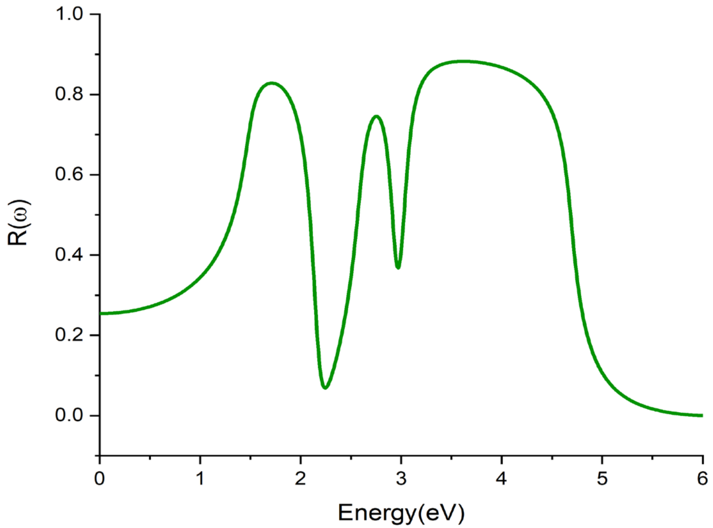
S. Saha, T. Sinha, and A. Mookerjee, Phys. Rev. B 62, 8828 (2000).



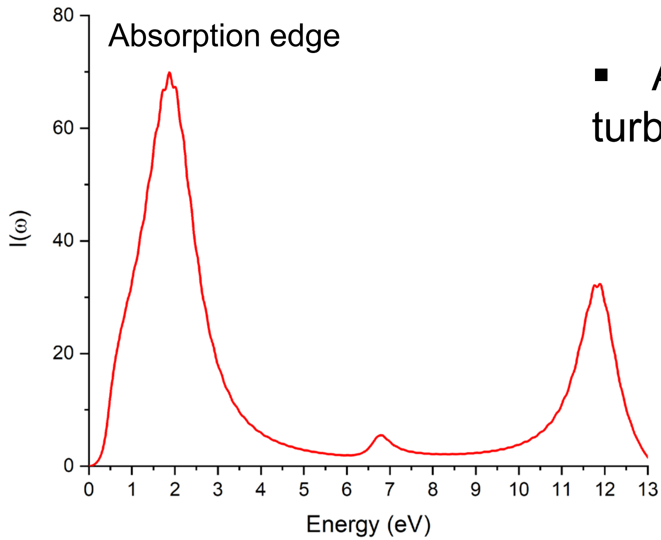
- Refractive index n(ω)



- Extinction coefficient k(ω)



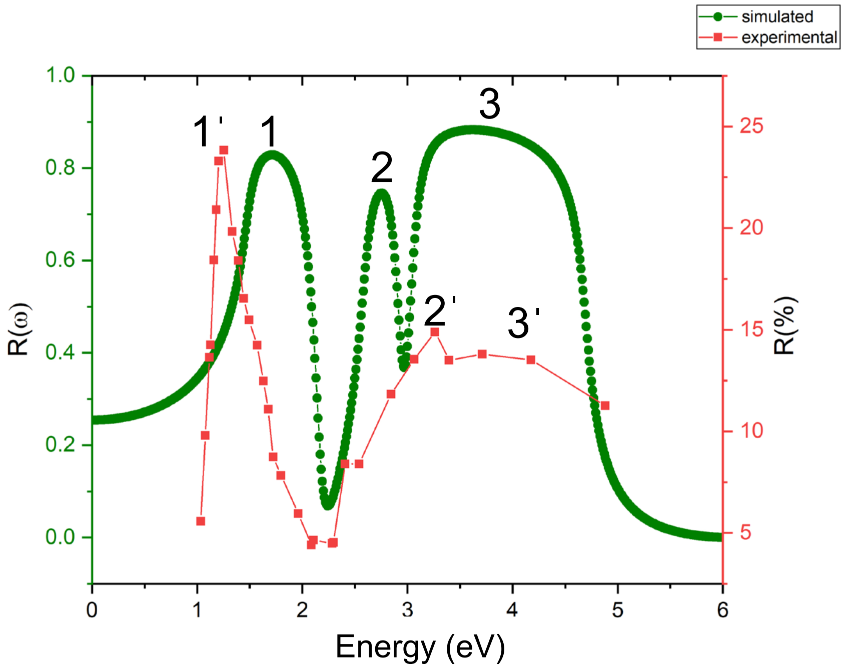
- Reflectivity R(ω)

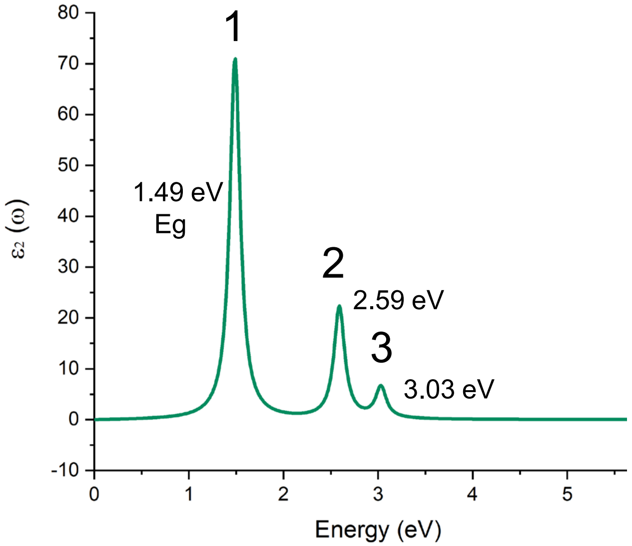


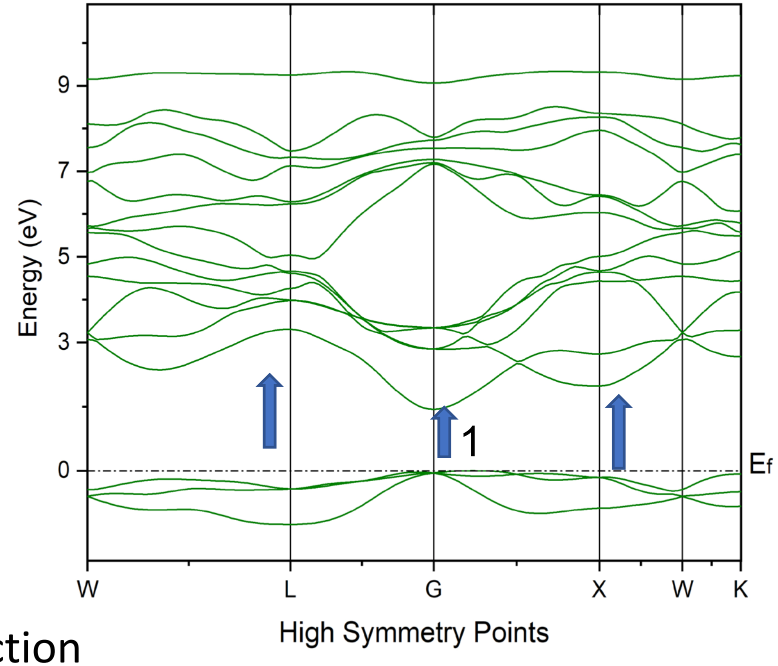
- Absorption spectra calculated by using turboTDDFT package in Quantum Espresso.

- TDDFT Absorption spectra I(ω)

K_2CsSb Optical Properties



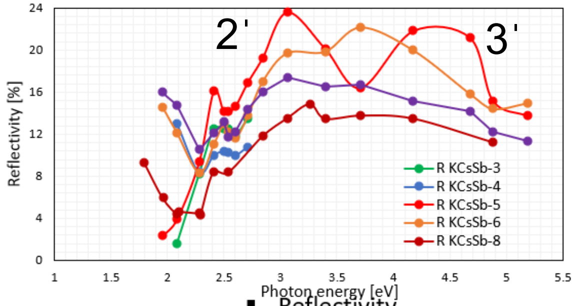




- Fig 1: Reflectivity comparison
- Fig 2: Imag. part of the dielectric function

* Here the transmittance value of viewport is not included in experimental reflectivity data

- The behavior of the experimental and simulated Reflectivity curves are quite similar.
- The fundamental absorption peak (first peak) in fig: 1 & 2 is due to the optical transition between G-G.
- From the above observation of the experimental reflectivity curve, we estimated the bandgap of KCsSb is 1.1-1.2 eV.



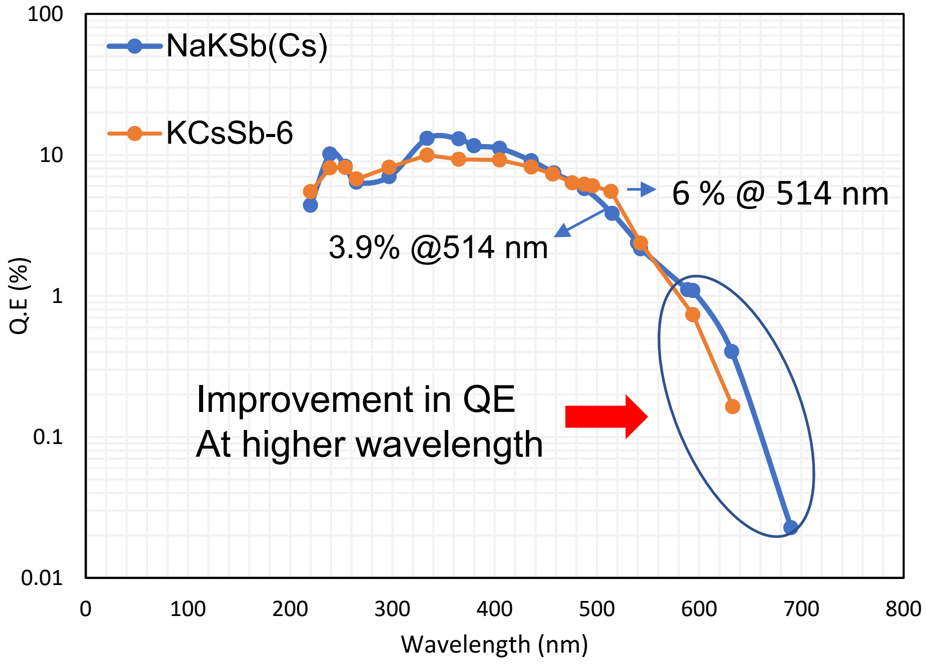
- Reflectivity

* Here the transmittance value of viewport is not included in QE and reflectivity data

NaKSb(Cs)

- Total of two NaKSb(Cs) cathodes has prepared in the R&D preparation chamber.
- During COVID, NaKSb(Cs)#2 has survived almost **two and half years** in the preparation chamber (Pressure 10 ¹¹⁻ mbar) .
- So far we have achieved, max Q.E ~ 3.9 % @514 nm with a peak Q.E of 13%@ 334 nm.
- From the comparison of the spectral response behavior of a KCsSb

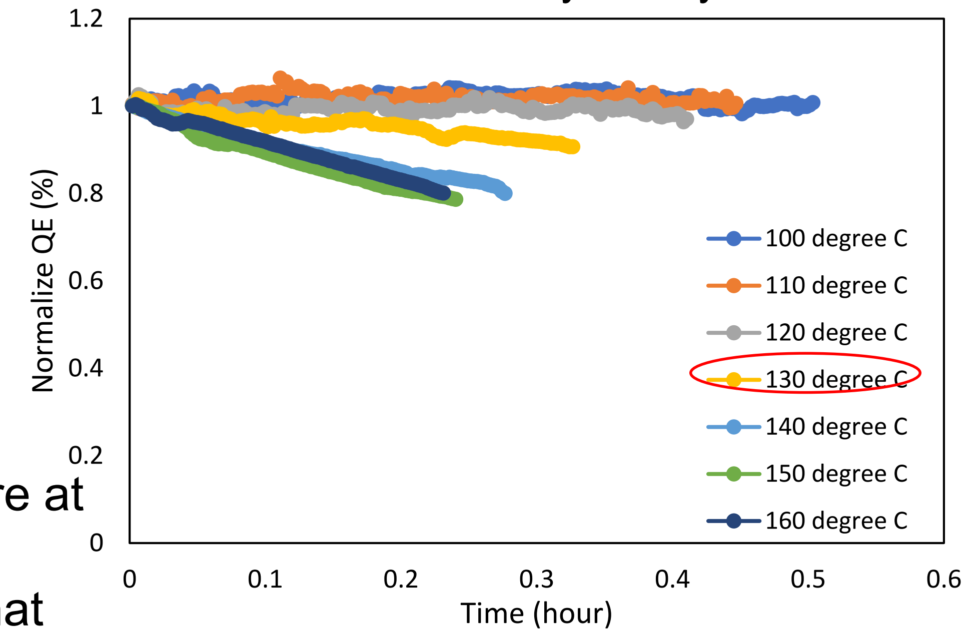
(Sb = 5 nm) and NaKSb(Cs) photocathode reveals that a slightly improved QE value for the NaKSb(Cs) cathode at a higher wavelength (typically the response to red laser light).

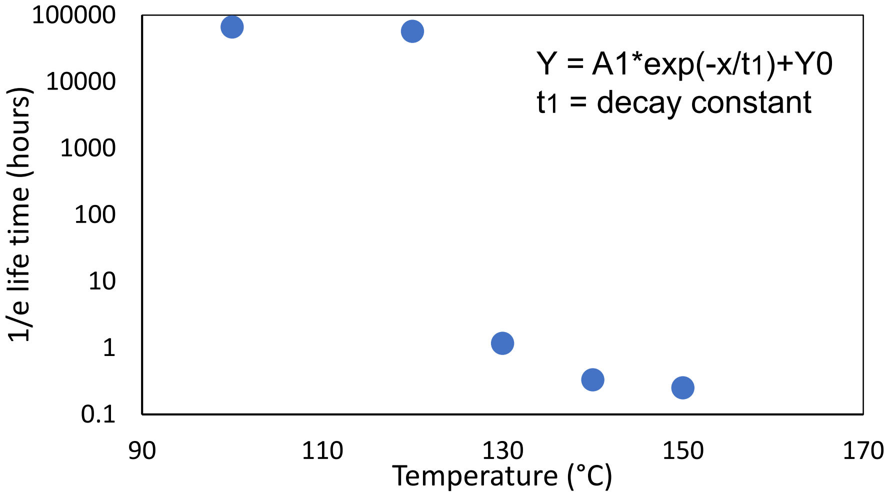


▪ Cathode degradation study (NaKSb(Cs) #2)

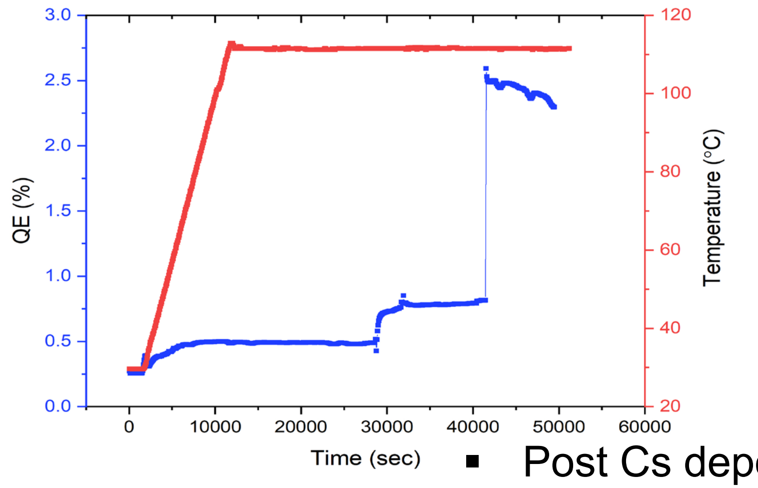
- Lifetime measurement of NaKSb(Cs) #2 cathode at various substrates the temperature was carried out.
- The cathode was kept at a particular temperature of about 30 to 60 minutes.
- The QE of the cathode is almost constant up to 120 °C and started to degrade at about **130 °C.**
- During the degradation study, the QE of the cathode was reduced from 1.2% to almost 0.3 % at 543 nm.
- Due to the low melting point of Cs (i.e., 28.44 °C), we assumed here at the elevated temperature we presumably lost most of **Cs**.
- So, we decided to deposit a few nm of Cs on it and we observed that QE was increased from **0.3 %** to **2.5%** at 543 nm.

- Decay history





- Life time analysis



- Post Cs deposition

Summary & Future plan

- Total of 8 cathodes produced with sequential deposition with QE @514 nm is recorded 5-9 % at 515 nm in the R&D chamber.
- 3 cathodes have been produced with sequential deposition with QE @514 nm is recorded 4-8 % for the test in RF gun at PITZ.
- The results in terms of QE, thermal emittance, and response time are very promising. However, the limiting factors are high dark current and short operational lifetime.
- Post-usage analysis shows different levels of oxidation of the cathode films.
- Refracting index has been calculated for Mo (values are similar to the reference value) and overall estimated the refracting index value for KCsSb cathode at different wavelengths of light.
- DFT calculations have been done by using QUANTUM ESPRESSO for KCsSb cathode.
- Optical properties (reflectivity, refracting index, extinction coefficient, etc.) have been calculated and compared with the experimental value
- NaKSb(Cs) cathode :
  - Total of two NaKSb(Cs) cathodes has prepared with max Q.E ~ 3.9 % @514 nm with a peak Q.E of 13%@ 334 nm
  - Study on NaKSb(Cs) #2 shows that these photocathodes can **survive** up to **several years** inside the vacuum chamber.
  - NaKSb(Cs) photocathode can survive up to **120 °C** and thereafter it starts to degrade.

❑ Future Plans:

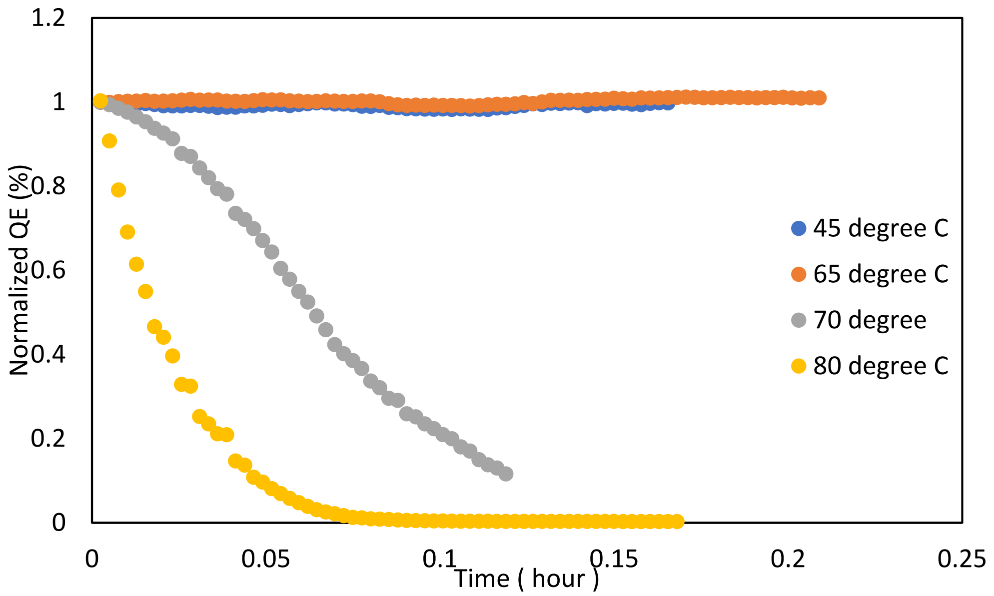
- Cathode degradation studies at different setup temperatures, gases, etc of new cathodes.
- Improve and further optimize the KCsSb cathode recipe (dep. rate, temperature, heterostructure, etc.)
  ➡ to improve the cathode's stability and QE.
- Develop a reproducible growth procedure for NaKSb(Cs) photocathode and study its stability & performance.
- Surface characterization study.

Thank You!

Backup Slides

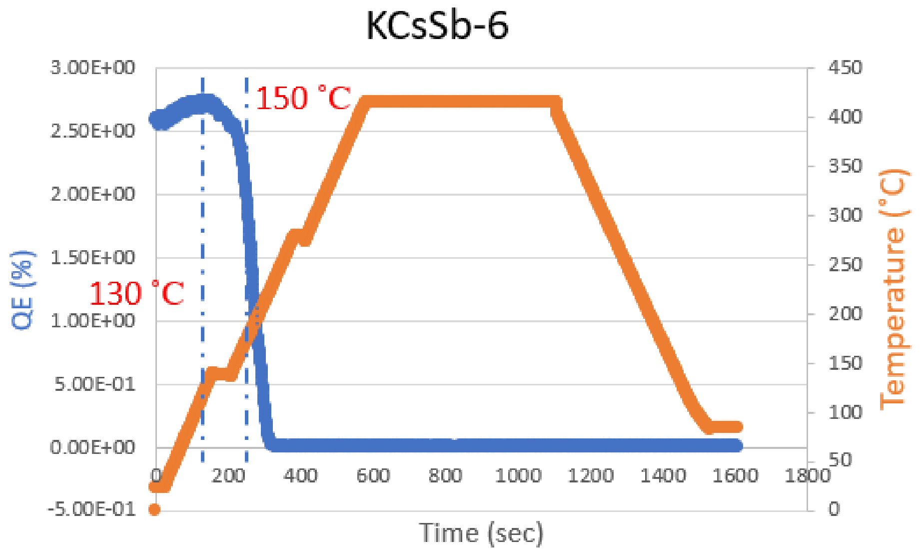
Cathode degradation study (KCsSb #8)

- The KCsSb #8 cathode was not complete (due to low Cs), compared to other thin cathodes.
- However the other previous photocathodes were degraded at about 130 °C .



Cathode	Sb [nm]	K [nm]	Cs [nm]	K/Sb	Cs/Sb	QE @514 nm
KCsSb-4	90 °C, 5 ± 0.9	90 °C, 41± 0.1	90 °C, 106 ± 0.5	8.2	21.2	5.54 %
KCsSb-6	90 °C, 5 ± 0.9	120 °C, 32 ± 0.1	90 °C, 117 ± 0.5	6.4	23.4	6.54 %
KCsSb-7	90 °C, 5 ± 0.9	120 °C, 34 ± 0.5	110 °C, 121 ± 0.5	6.8	24.2	7.11 %
KCsSb-8	90 °C, 5 ± 0.9	130 °C, 41± 0.5	120 °C, **35 ± 0.5**	8.2	7	**8.83 %**

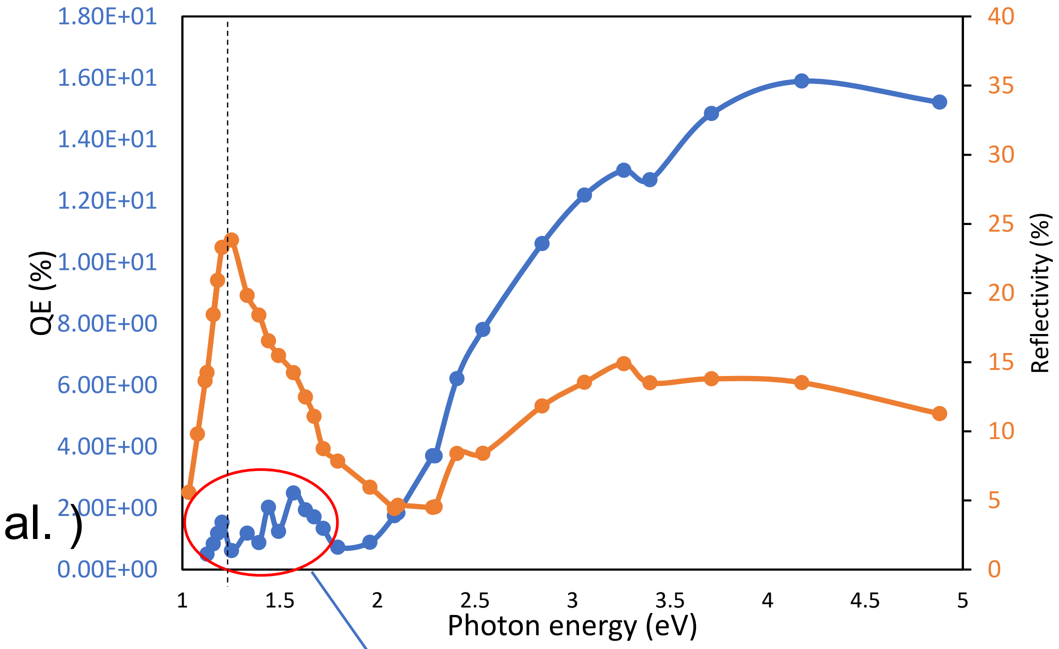
- Summary of cathode growing parameters of thin cathodes



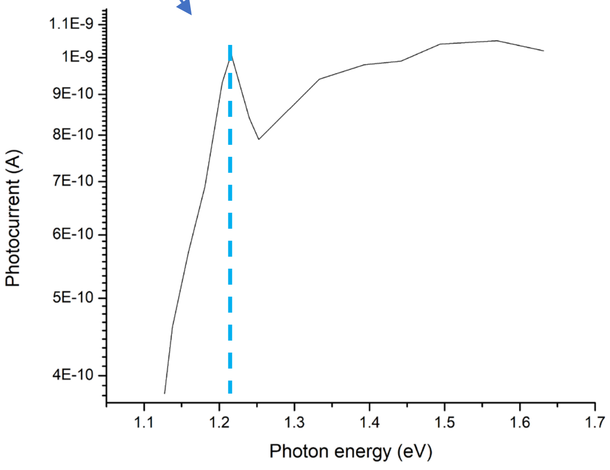
KCsSb #8

- A full spectral response + reflectivity has been measured for KCsSb #8 (from 1210 nm to 254 nm) .
- Both **reflectivity and the QE** sharply decreased at **1.2 eV**.
- Through photoconductive measurement, we estimate the **band gap** of the photocathode is **1.2 eV.**
- From the spectral response we also estimate the photoemission threshold is **1.9 eV.**
  (Reference Eg+Ea = 1.9 eV , Eg= 1.2 eV by Gosh & Verma et al.)

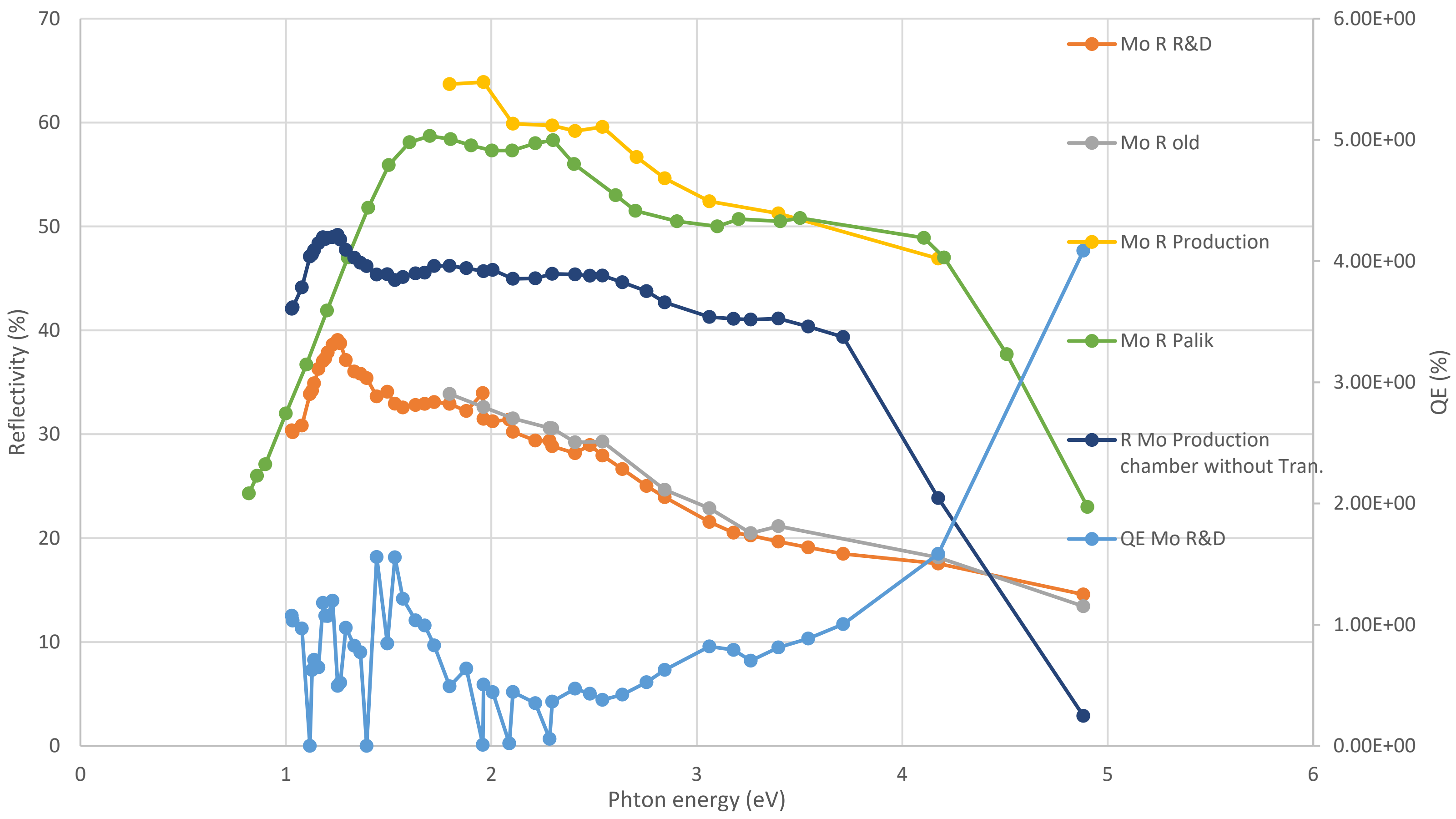
- Full Spectral response



* Here the transmittance value of viewport is not included in QE and reflectivity data



Mo (R&D)

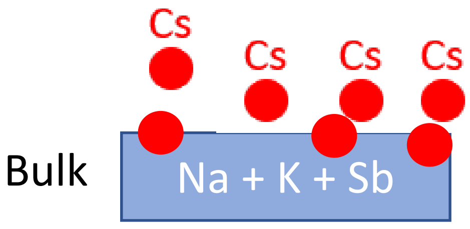


Reflectivity (%)
QE (%)
Phton energy (eV)
0
10
20
30
40
50
60
70
0
1
2
3
4
5
6
0.00E+00
1.00E+00
2.00E+00
3.00E+00
4.00E+00
5.00E+00
6.00E+00
Mo R R&D
Mo R old
Mo R Production
Mo R Palik
R Mo Production chamber without Tran.
QE Mo R&D

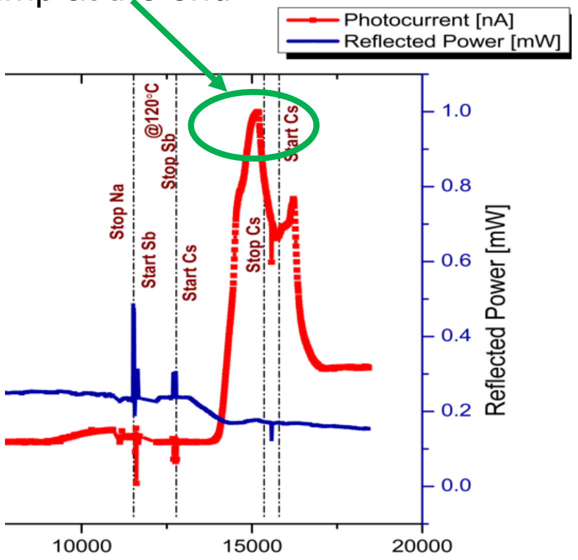
Reaction of Cs with different bulk agent

KCsSb Cathode

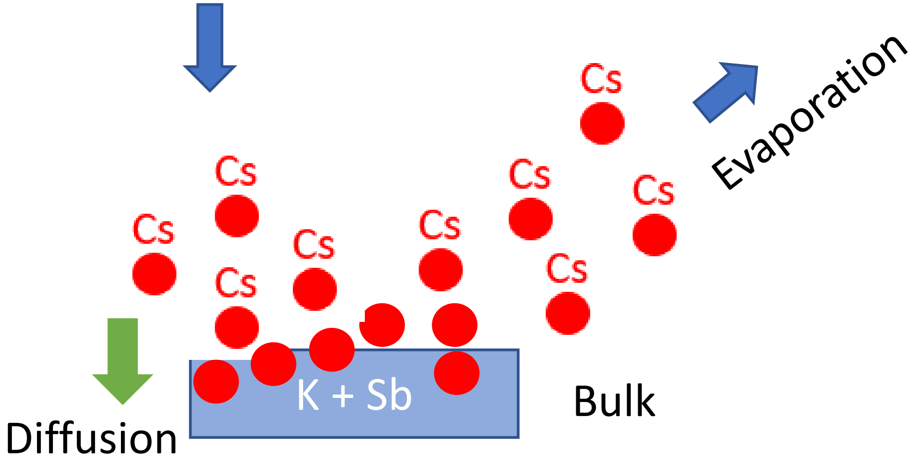
NaKSb(Cs) Cathode



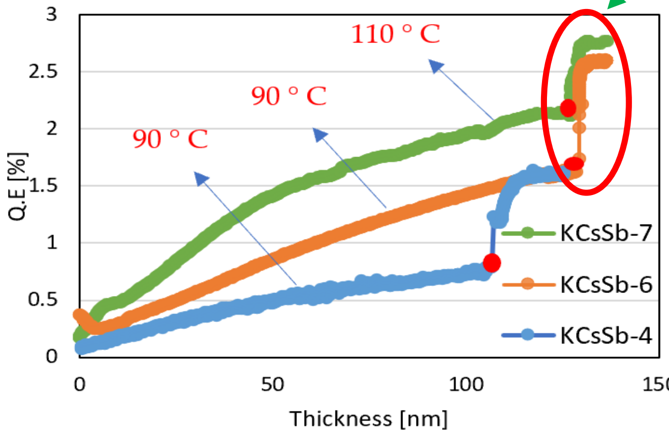
No jump at the end



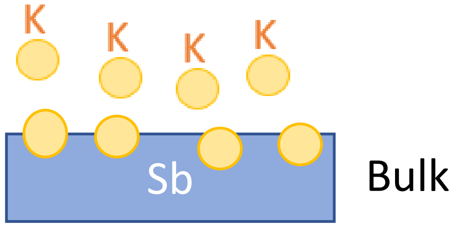
Incoming Cs



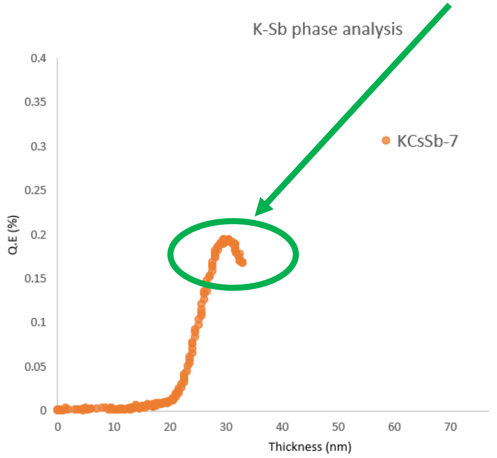
jump at the end



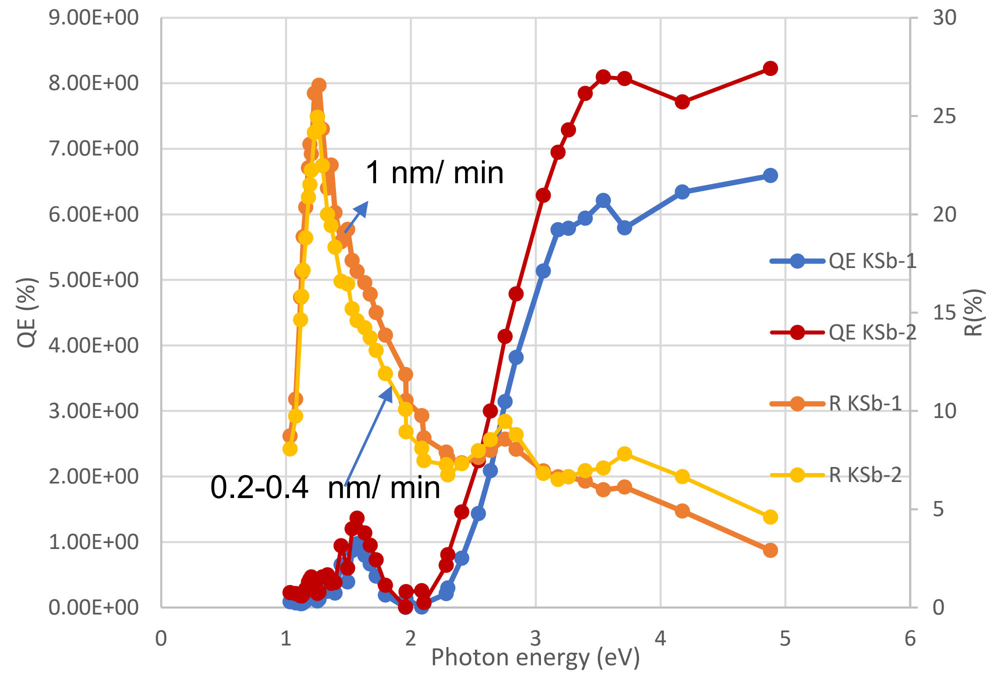
K3Sb Cathode

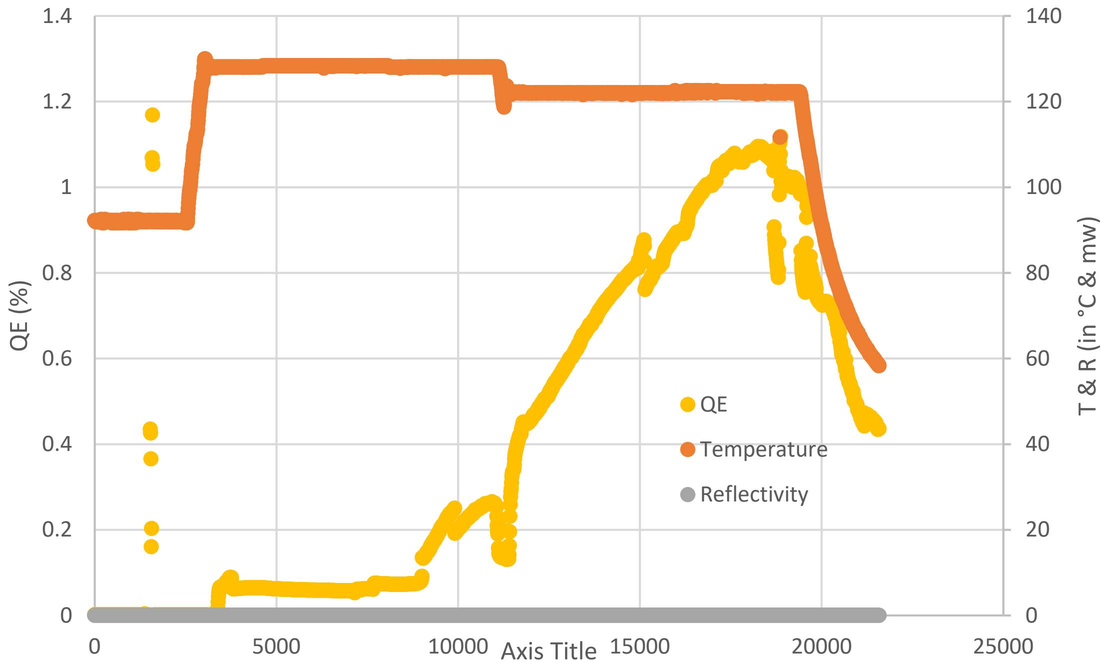


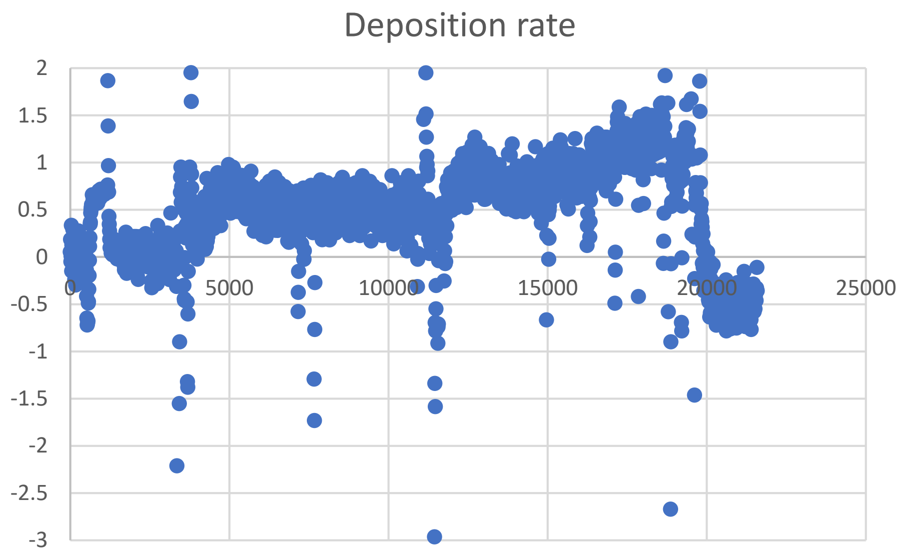
No jump at the end



KSb #02





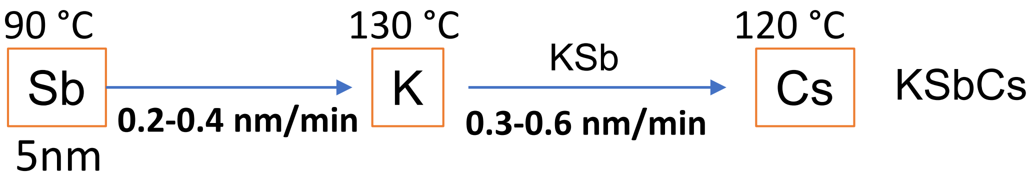


QE @543 nm: 1% (final for KSb-2)

Low deposition rate can enhance QE for K-Sb

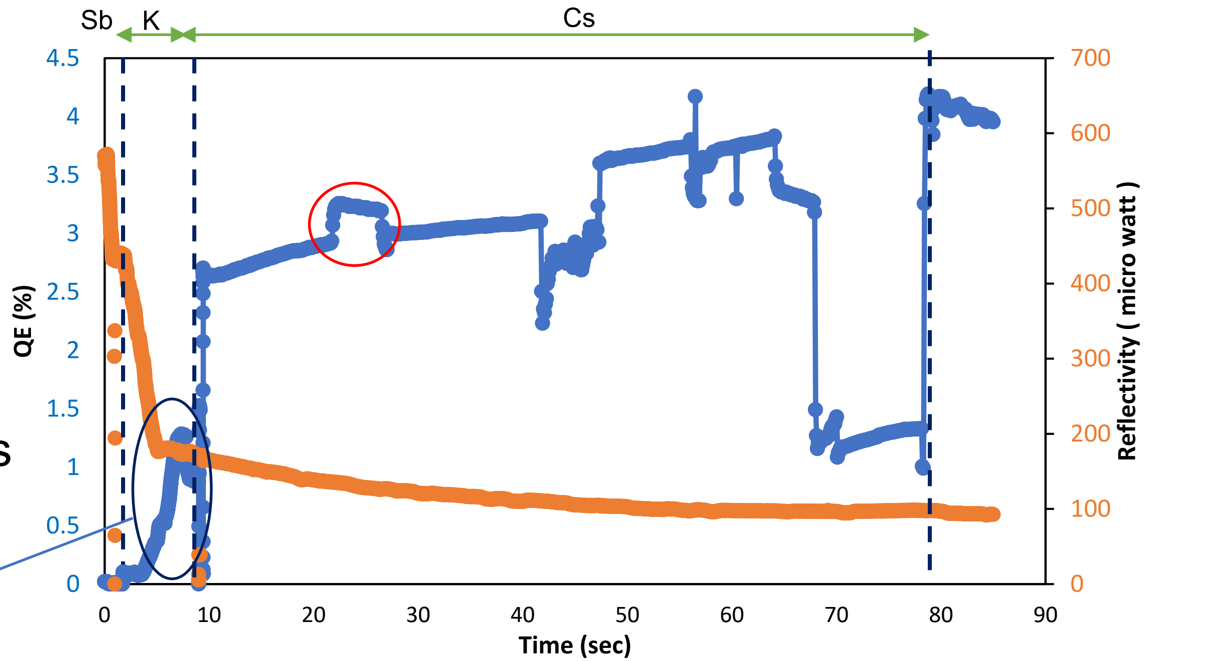
"New" KCsSb #8 cathode

- KCsSb #8 has grown in the R&D preparation chamber through sequential deposition method.

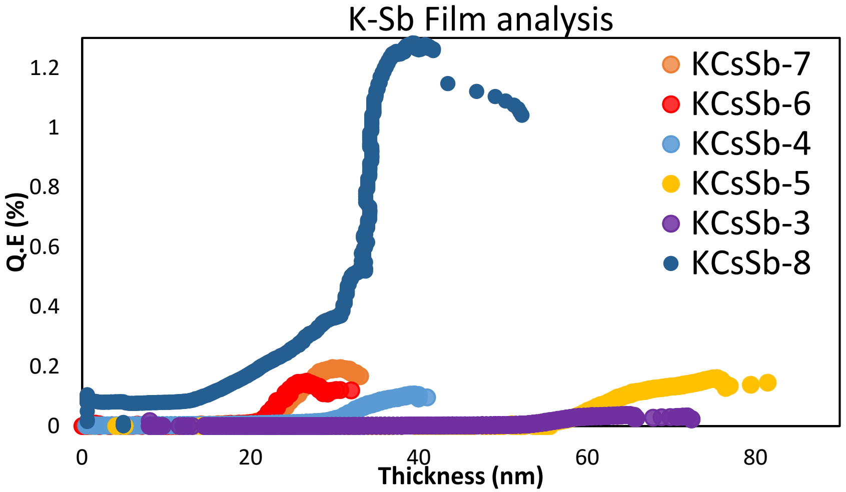


- Deposition rate has been changed compared to previous methods (i.e., 1 nm/min) during K and Cs deposition.
- Due to little elevated temp. 130 °C (before 120 °C) and slow deposition rate (**0.2-0.4** nm/min), which helped to improve the **QE** (record high ~ **1.2 % at 543 nm**) for **KSb** film.
- Unfortunately, the Cs source was completely empty during the Cs deposition, so we observe a disturbance in the QE curve.
- The final QE of KCsSb #8 is
  - **5.1** % @543 nm (compare to 3.07 % for thin cathodes)
  - **8.84** %@ 515 nm (compare to 6.88 % for thin cathodes

  (the QEs are estimated by including the transmittance of the viewport)



- Photocurrent and reflectivity history (not include transmittance of the viewport



* Here the transmittance value of viewport is not included in QE data