Development of Multialkali antimonides photocathodes for high brightness photoinjectors

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Contents:

- Multi-alkali antimonides photocathodes development
- Why the Optical properties are important?
- Experimental results
- Photocathode recipes
- Density Function Theory (DFT) study (K_2CsSb , K_3Sb)
- Summary & Future plan

Multi-alkali antimonides photocathodes development

- **Alkali based Photocathodes** (K_2CsSb, Na_2KSb, Cs_3Sb,….) have shown great potential in **low gradient** guns (<20 MV/m)
  - Cornell DC gun, BNL SRF gun
  - **>1 % QE** at green wavelengths, **0.5 - 0.6** mm.mrad/mm **thermal emittance**
  - High average current
  - Improves cathode laser efficiency and shaping

- **DESY** collaborates with **INFN LASA** to explore multi-alkali photocathode performance in **high-gradient** guns
  - INFN LASA, Italy, develops cathode recipe and production
  - Photoinjector test facility at DESY Zeuthen site (PITZ) tests cathodes in a high-gradient RF gun.

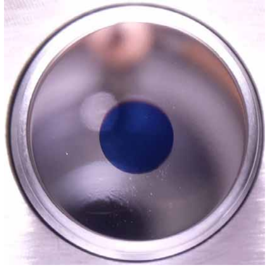
- In the **R&D stage** (produced a total of 8 cathodes), **a reproducible recipe has been achieved** for the **KCsSb** compound with a maximum QE of ~ 9% @ 515 nm [1].

- **3 KCsSb** photocathodes were prepared at INFN LASA in July 2021 and successfully tested at PITZ **RF gun** [2].
  - ✓ High QE (**4-8 %** at 515 nm)
  - ✓ Thermal emittance (**0.6** mm.mrad/mm) (lower than Cs_2Te)
  - ✓ Response time (preliminary results show **< 100 fs**)
  - Higher dark current than Cs_2Te
  - Short operational lifetime (~48 hours)

✓ Thin cathodes: Sb 5 nm

✓ Thick cathodes: Sb 10 nm


112.1 (thin)


147.1 (thick)

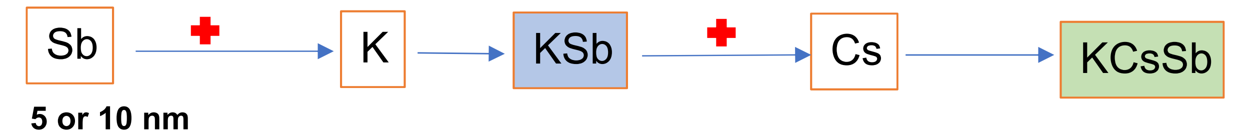

123.1 (thin)

[1] Mohanty SK, Krasilnikov M, Oppelt A, Stephan F, Sertore D, Monaco L, Pagani C, Hillert W. Development and Characterizati of Multi-Alkali Antimonide Photocathodes for High-Brightness RF Photoinjectors. Micromachines. 2023; 14(6):1182. https://doi.org/10.3390/mi14061182 .

[2] S. Mohanty, "Development and Test Results of Multi-Alkali Antimonide Photocathodes in the High Gradient RF Gun at PITZ", Proc. FEL2022, Trieste. doi:10.18429/jacow-fel2022-tup04

Multi-alkali antimonides photocathodes development

- **To improve + optimize cathode recipe** :
  - ✓ Two new cathodes grown in the new “production” system.
    - One **thick** (Sb = 10 nm) (**#137.2**)
    - One **thin** (Sb = 5 nm) (**#137.3**)

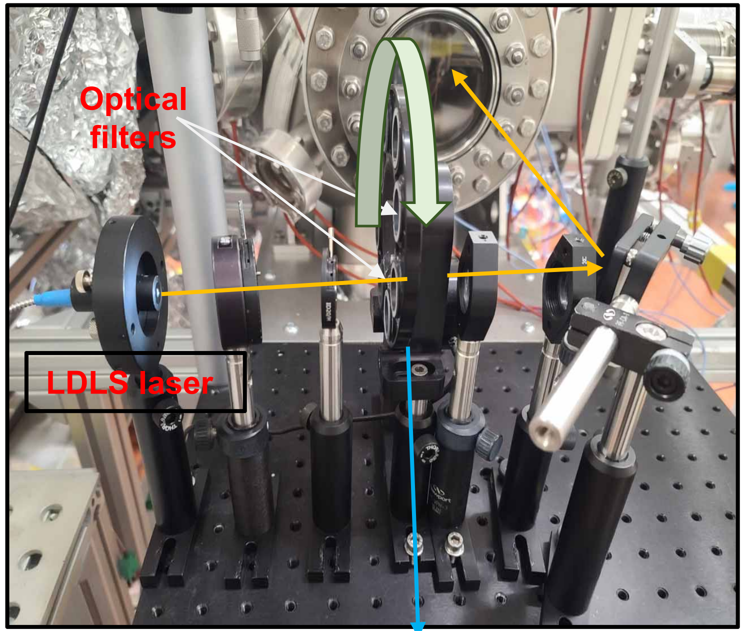


Sb + → K → KSb + → Cs → KCsSb

5 or 10 nm

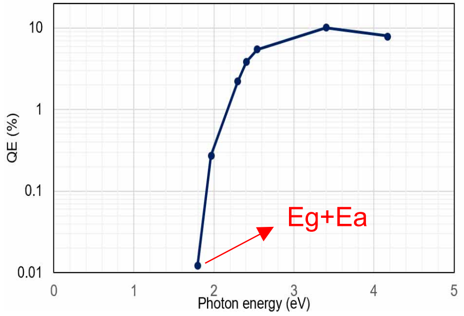
Sequential deposition method

“Multi-wavelength” Optical Diagnostics used during cathode deposition



Motorized filter wheel (housing **8 optical filters** with different wavelengths)

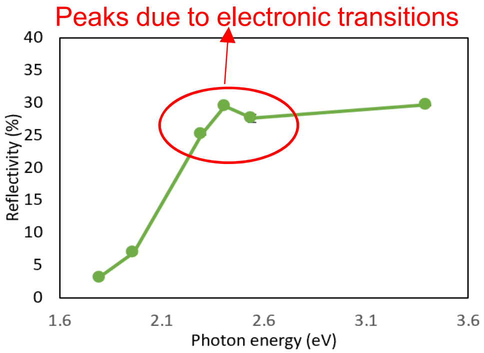
- ➢ Real-time **Spectral response**
  - ✓ Continuous **tracking** of the **Eg+Ea** value in real time.
    - Identify the formation of new compounds (transition from **Sb** to **KSb** and then to **KCsSb**)
  - ✓ Reaction Kinetics
    - revealing reaction rates and intermediate stages.



Eg = band gap
Ea = electron affinity

➕

- ➢ Real-time **Spectral reflectivity**
  - ✓ Optical Characterization
    - provide insights into the energy **band structure** and electronic transitions within the compound



Why the Optical properties are important?

- Determining the electronic structure of a material through optical measurements, such as **spectral reflectivity** and **spectral response**, is considered an **indirect method** (because it relies on interpretations and correlations rather than direct measurement of electronic states)**.**

Optical Properties

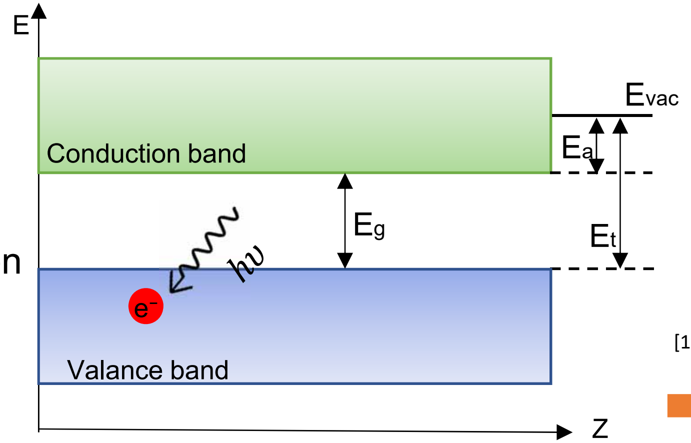
Spectral reflectivity

Polarisation: $P(\omega) = \epsilon_0\, \tilde{\chi}(\omega)\, E(\omega)$

Complex dielectric function: $\tilde{\epsilon}(\omega) = 1 + \tilde{\chi}(\omega)$

Real part: $\epsilon'(\omega) = \epsilon_1 = 1 + \tilde{\chi}'(\omega)$

Imaginary part: $\epsilon''(\omega) = \epsilon_2 = \tilde{\chi}''(\omega)$ → Absorption



E_g = Band gap

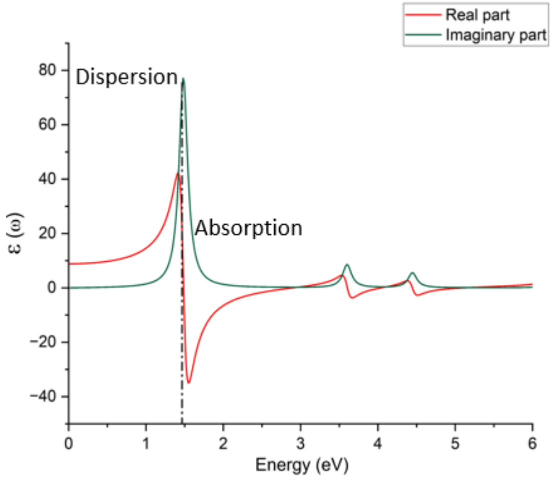
E_a = Electron affinity

E_t = Photoemission threshold

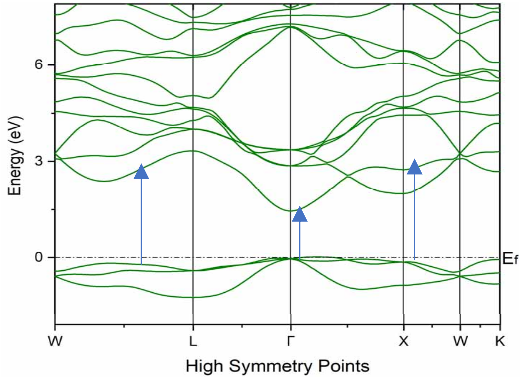
Maxwell model [1]

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

[1] F. Wooten, "Chapter 1 - introduction," in Optical Properties of Solids,F. Wooten, Ed. Academic Press, 1972, pp. 1–14.

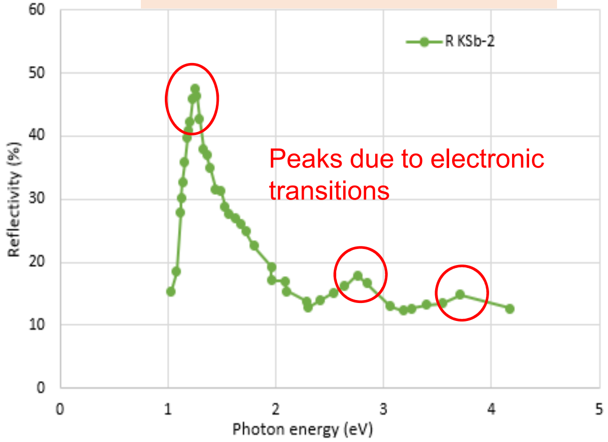


Dielectric function



Band structure

Spectral reflectivity

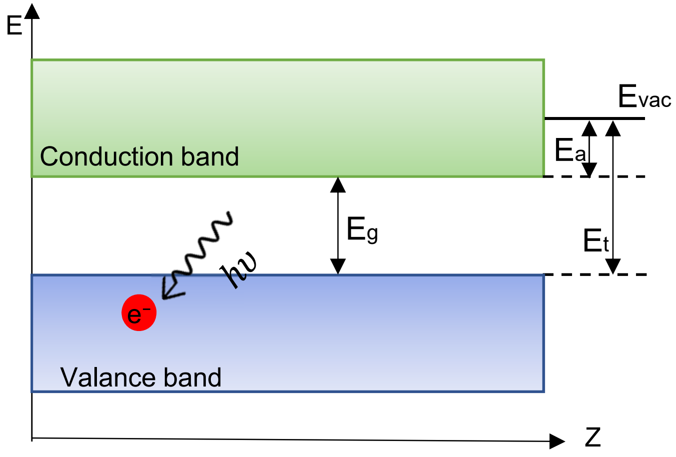


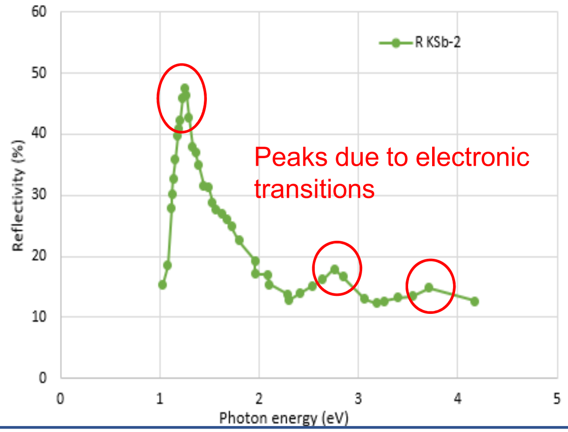
Peaks due to electronic transitions

Why the Optical properties are important?

- Determining the electronic structure of a material through optical measurements, such as **spectral reflectivity** and **spectral response**, is considered an **indirect method** (because it relies on interpretations and correlations rather than direct measurement of electronic states)**.**

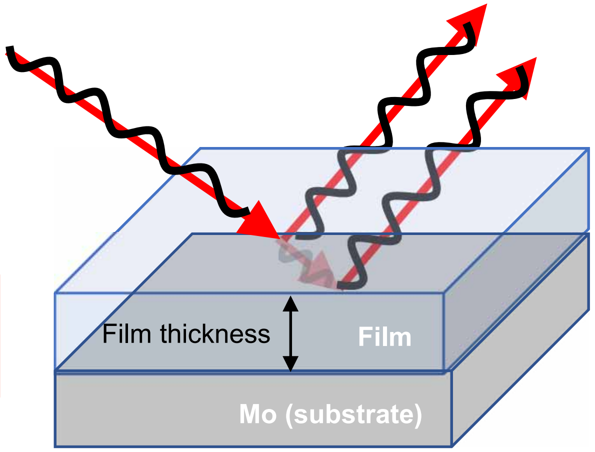
Optical Properties → Spectral reflectivity





Peaks due to electronic transitions

Not simplified to interpret directly



Interference effects due to thickness or the presence of multiple layers (such as thin films and substrates) Also, surface roughness, etc. can influence.

- Interpretation of optical measurements requires **theoretical models** and assumptions about the underlying electronic structure.
- The **dielectric function** (Kramer's-Kronig equation) is used to calculate the optical properties of the material.

Quantum Espresso software package and its postprocessing code epsilon.x

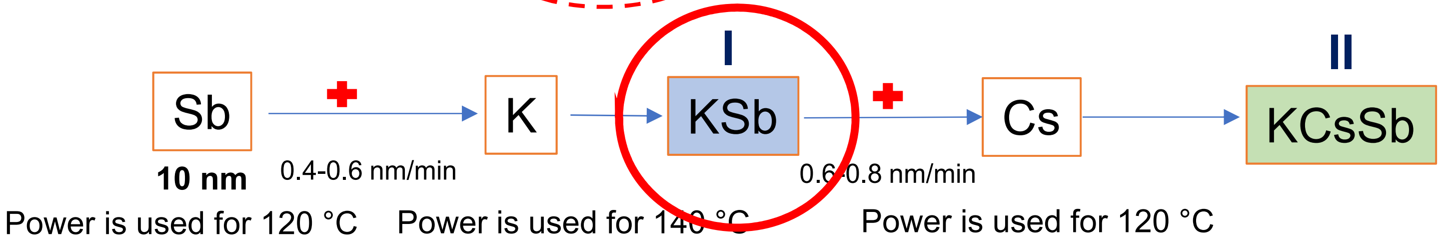
- Refractive index $n(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}-\varepsilon_1(\omega)}{2}\right]^{0.5}$
- Extinction coefficient $k(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}+\varepsilon_1(\omega)}{2}\right]^{0.5}$
- **Reflectivity**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

Optical properties like **reflectivity** etc. ⟷ Experimental reflectivity

Experimental results

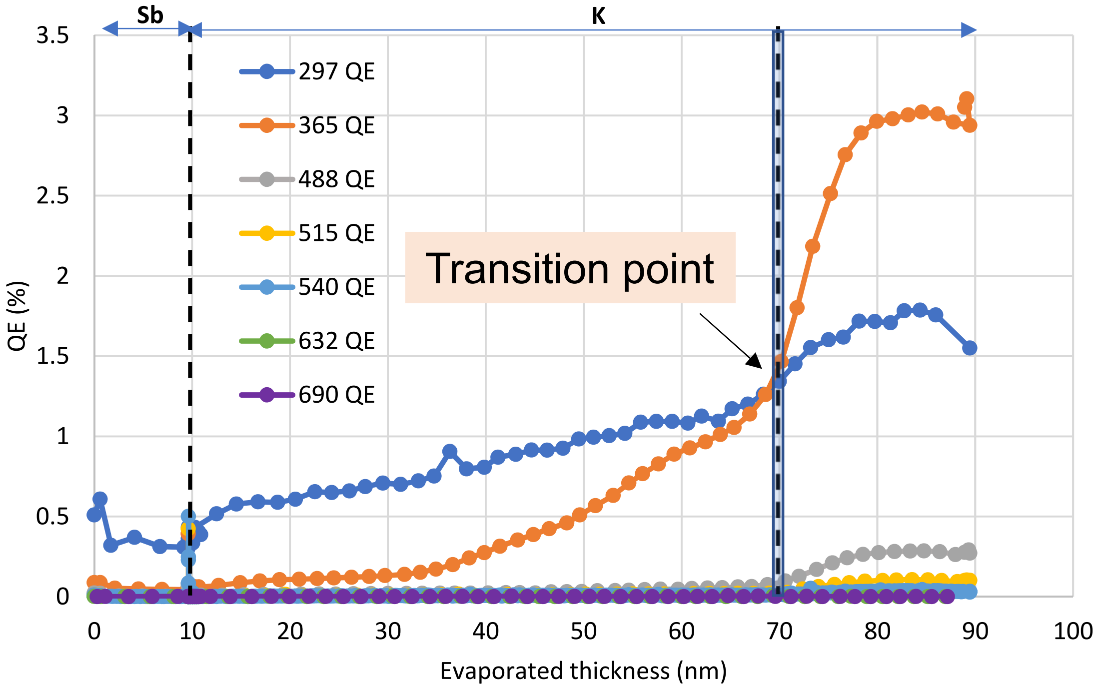
Cathode - 1

Cathode 137.2 (K+Sb+Cs), Sb = 10 nm (Thick)

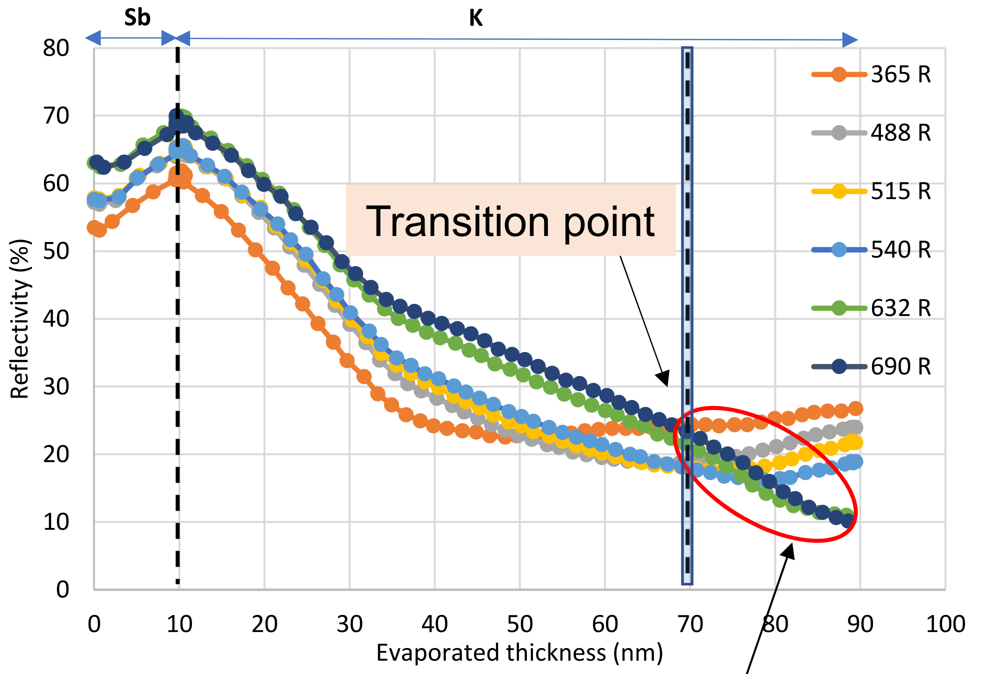


“Real-time” QE & Reflectivity curve 137.2 (KSb – Thick, Cathode-1)

- “Real-time” **QE** vs. evaporated **thickness** during Sb & K deposition



- “Real-time” **Reflectivity** vs. evaporated **thickness** during Sb & K deposition



- During **Sb** deposition, the **reflectivity** was **increased** (at 2 nm).
- After **70 nm** of **K** evaporation (transition point) [1,2], there is a **change** in **slope** in **real-time QE** (at all the wavelengths).
- After **70 nm** of **K** evaporation (transition point) [1,2], the **behaviour of reflectivity** for **red wavelengths** (**632** & **690** nm) has changed compared to other wavelengths.
- The rate of QE increase at 365 nm was higher compared to 297 nm only after 70 nm of K evaporation (e-e scattering!, if hν ≥ 2Eg).

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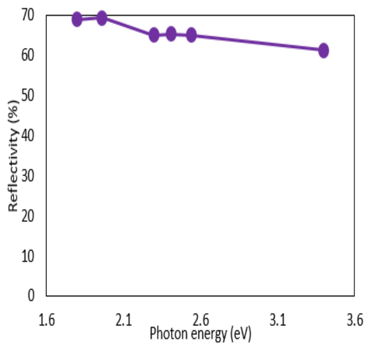
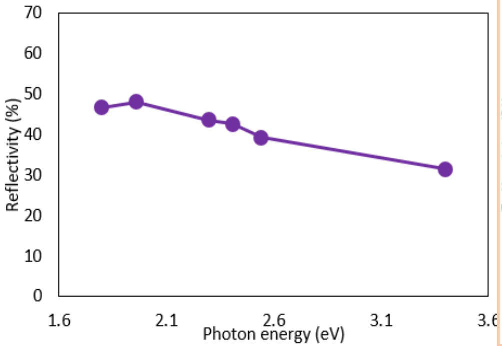
[1] Mohanty SK, Krasilnikov M, Oppelt A, Stephan F, Sertore D, Monaco L, Pagani C, Hillert W. Development and Characterization of Multi-Alkali Antimonide Photocathodes for High-Brightness RF Photoinjectors. Micromachines. 2023; 14(6):1182. https://doi.org/10.3390/mi14061182 .

[2] Ruiz-Osés, M.; Schubert, S.; Attenkofer, K.; Ben-Zvi, I.; Liang, X.; Muller, E.; Padmore, H.; Rao, T.; Vecchione, T.; Wong, J.; et al. Direct observation of bi-alkali antimonide photocathodes growth via in operando x-ray diffraction studies. APL Mater. 2014,2, 121101.

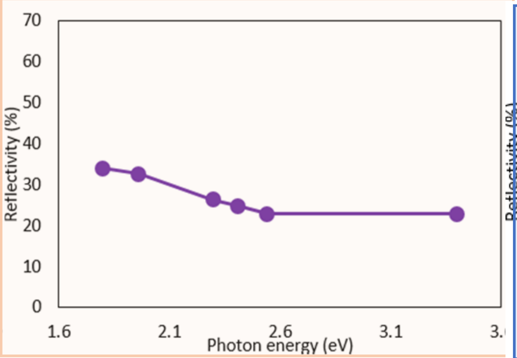
Real-time Reflectivity history during Sb+K deposition (137.2 KSb-Thick,Cathode-1

- "Real-time" **Reflectivity** vs. **Photon energy (spectral reflectivity)**

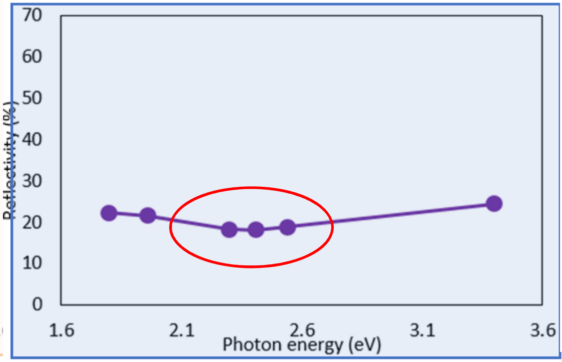




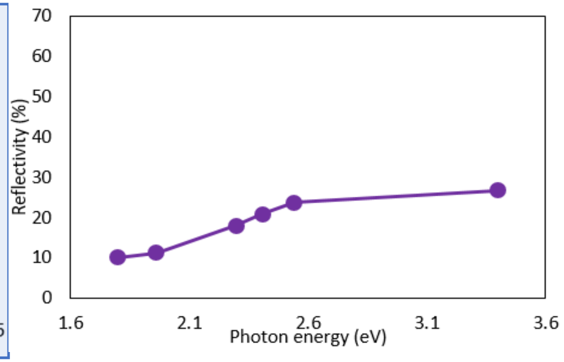
@30 nm of K



@50 nm of K



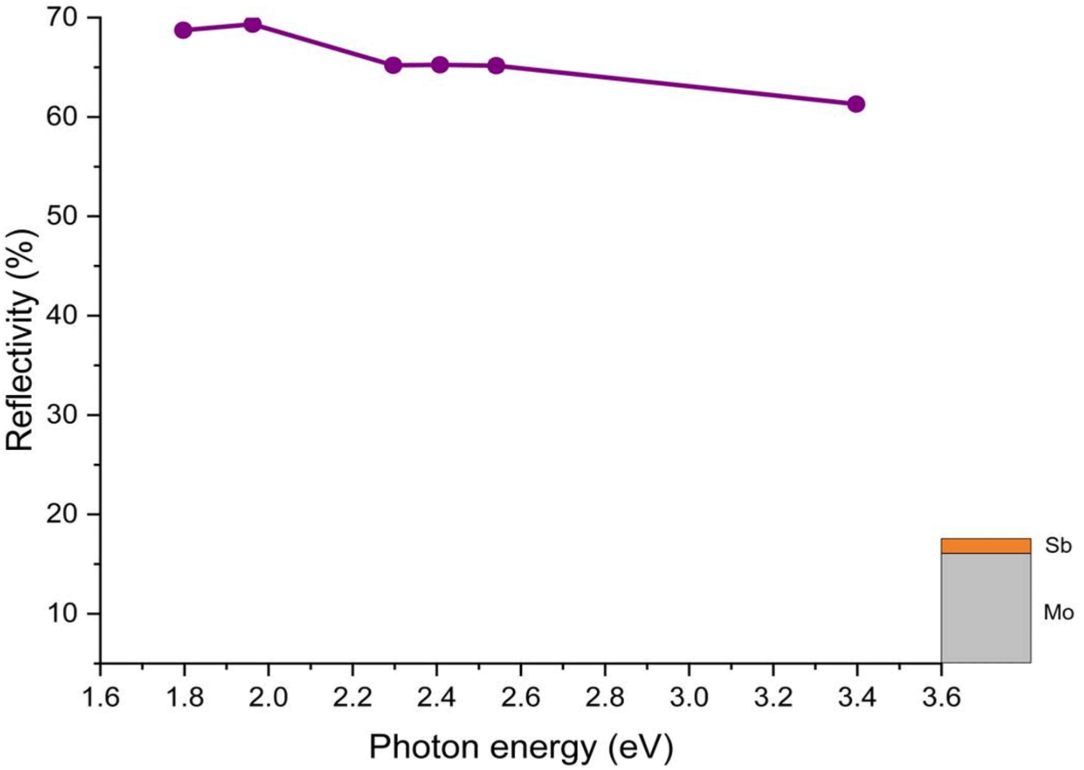
@70 nm of K



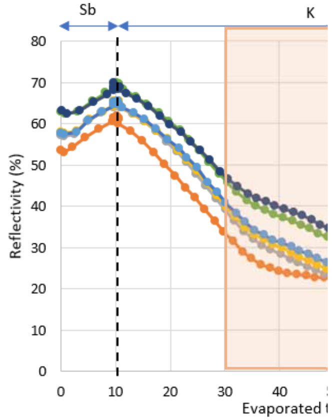
@88 nm of K

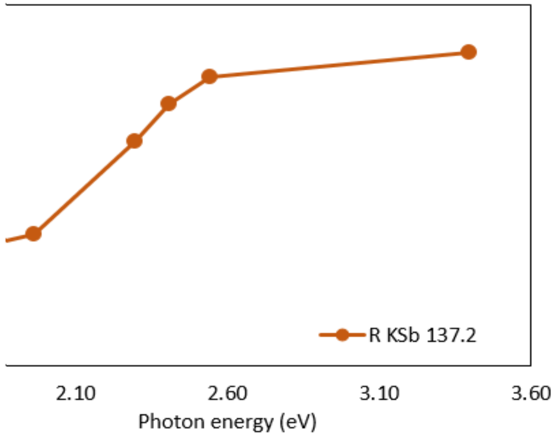
Sb deposition

R(%) history during Sb+K deposition (137.2 KSb-Thick)



- "Real-time" **Reflect**





Final Reflectivity

- Up to 30 nm of K evaporation, there wa
- From 30 to 50 nm of K evaporation, in t
  the shape of the spectral reflectivity (up
- At 70 nm of K evaporation (transition p

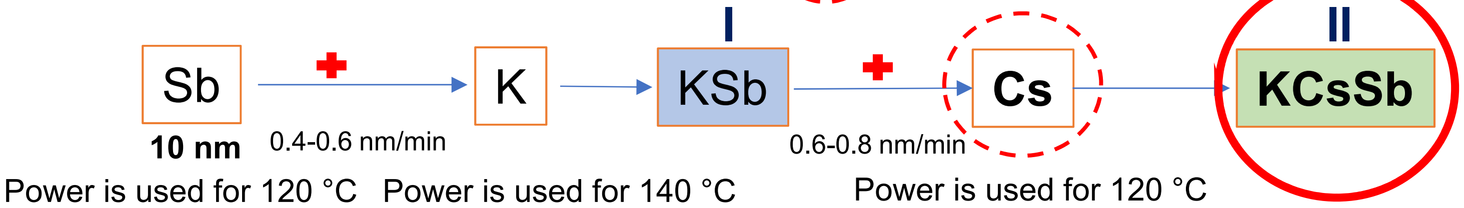
reased at all λ).

eflectivity observed at all λ, and

al reflectivity Fig. (upper plot).

Cathode - 1

Cathode 137.2 (K+Sb+Cs), Sb = 10 nm (Thick)



I

II

Sb → K → KSb → Cs → KCsSb

10 nm

0.4-0.6 nm/min

0.6-0.8 nm/min

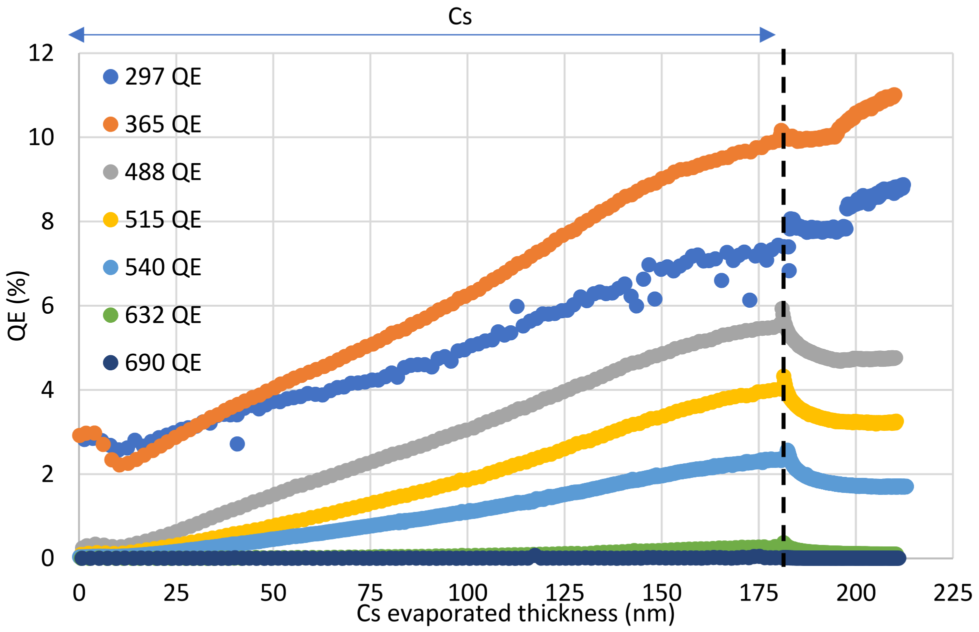
Power is used for 120 °C

Power is used for 140 °C

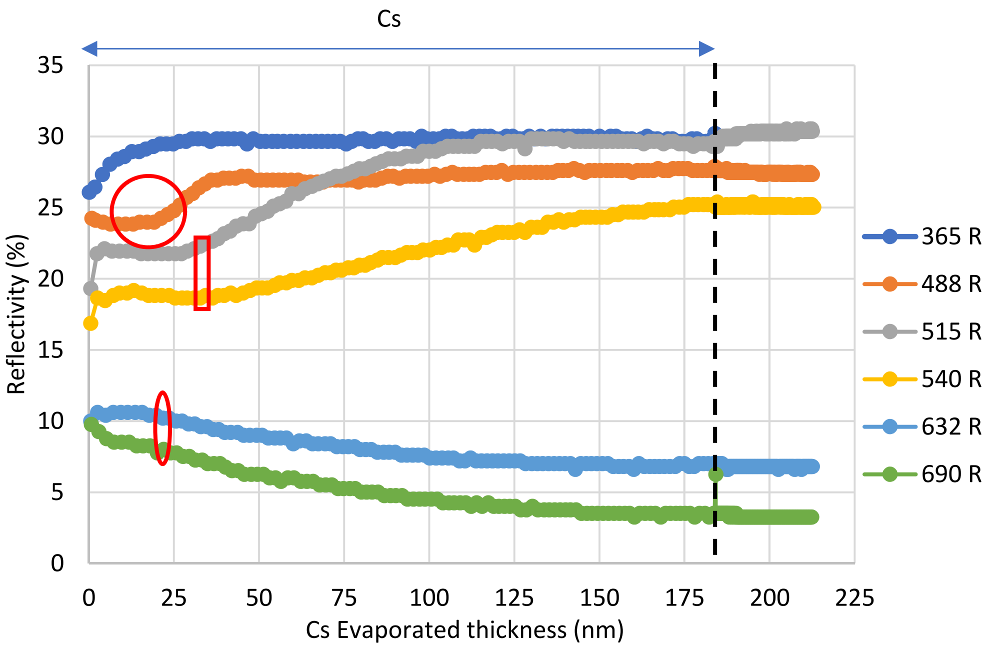
Power is used for 120 °C

"Real-time" QE & Reflectivity curve 137.2 (KSb+Cs – Thick, Cathode-1)

- "Real-time" **QE** vs. evaporated **thickness** during Cs deposition



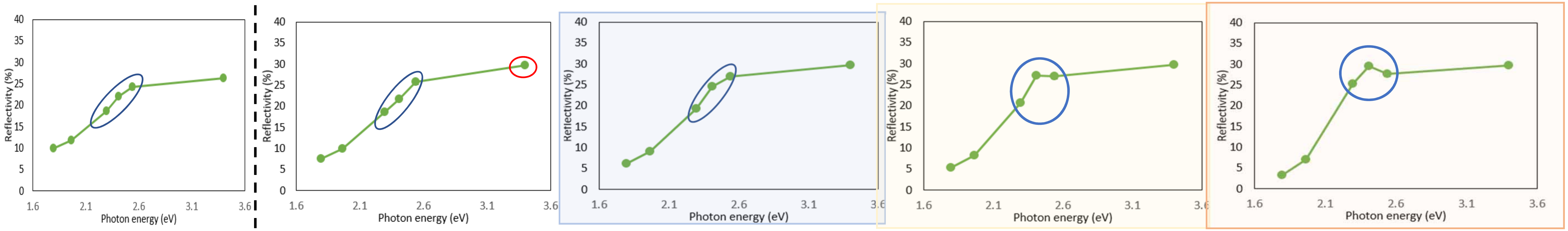
- "Real-time" **Reflectivity** vs. evaporated **thickness** during Cs deposition



- After **25 nm** of Cs evaporation, a **change in slope** in the **reflectivity curve** for **488 nm** has been noticed. At this point, the reflectivity of red wavelengths (632 & 690 nm) starts to decrease.
- After **38 nm** of Cs evaporation, the **reflectivity** for green wavelengths (**515 & 540 nm**) starts to **increase**.
- At the end of Cs evaporation, the QE of UV wavelengths (297 & 365 nm) stabilize initially and then start to increase, whereas for the rest of the wavelengths**,** it starts to decrease (excess Cs evaporation!)

- **Real-time Reflectivity history during KSb+Cs deposition (137.2 KCsSb-Thick, Cathode-1)**

- “Real-time” **Reflectivity** vs. **Photon energy (spectral reflectivity)**

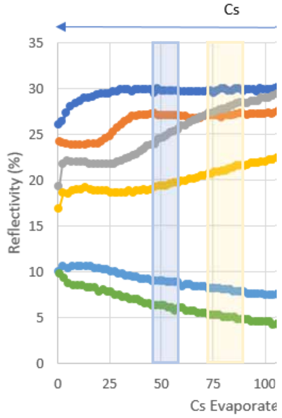


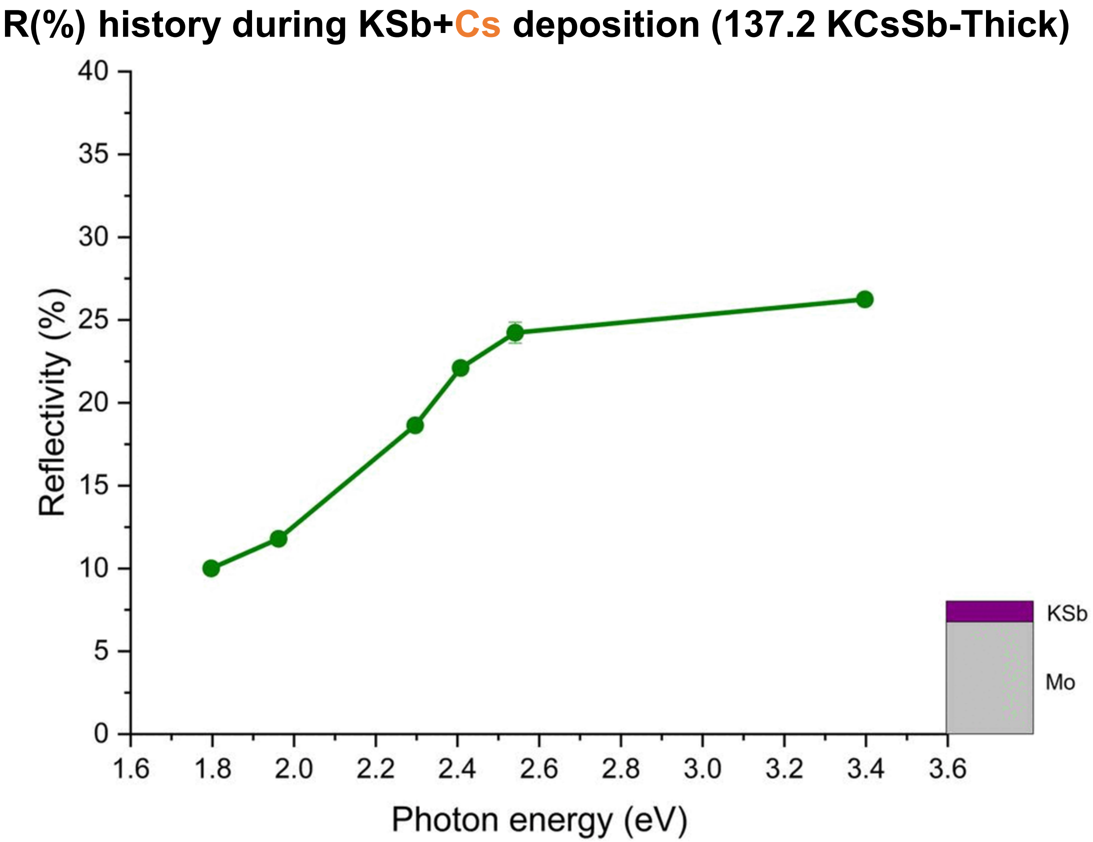
@30 nm of C

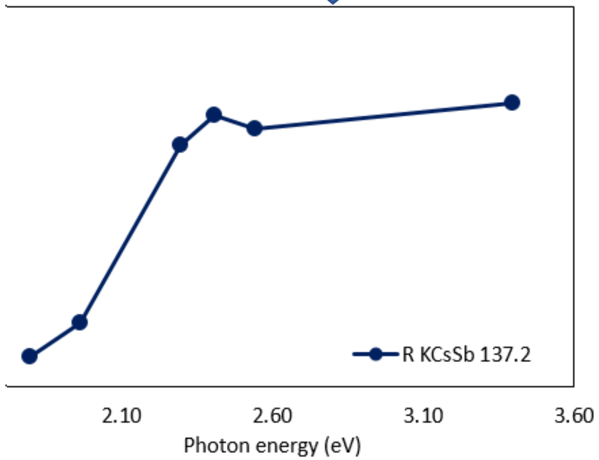
@183 nm of Cs

Sb + K deposition

- “Real-time” **Refle**







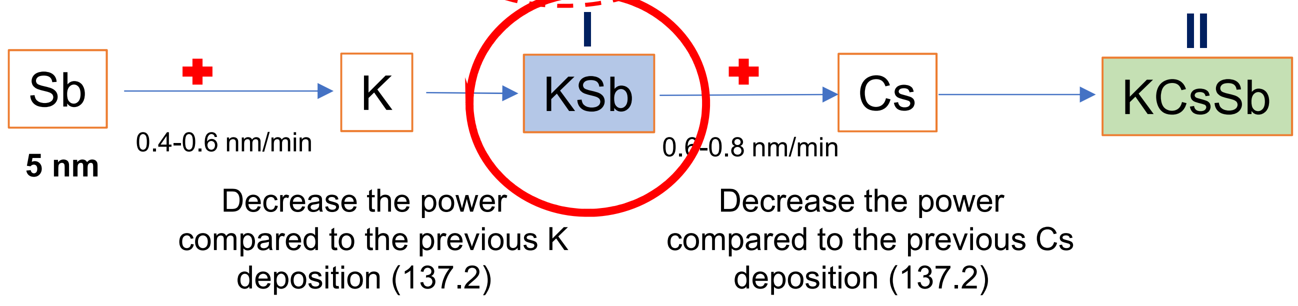
- Final Reflectivity

0 10 20 30 170 180 190 200 210 220

- Up to 30 nm of Cs evaporatic elength of light.
- Up to 76 nm of Cs evaporatic reased in the green wavelengths {515 (2.40 eV), 540 nm (2.29 t **2.40 eV** (515 nm).

Cathode - 2

Cathode 137.3 (K+Sb+Cs), Sb = 5 nm (Thin)

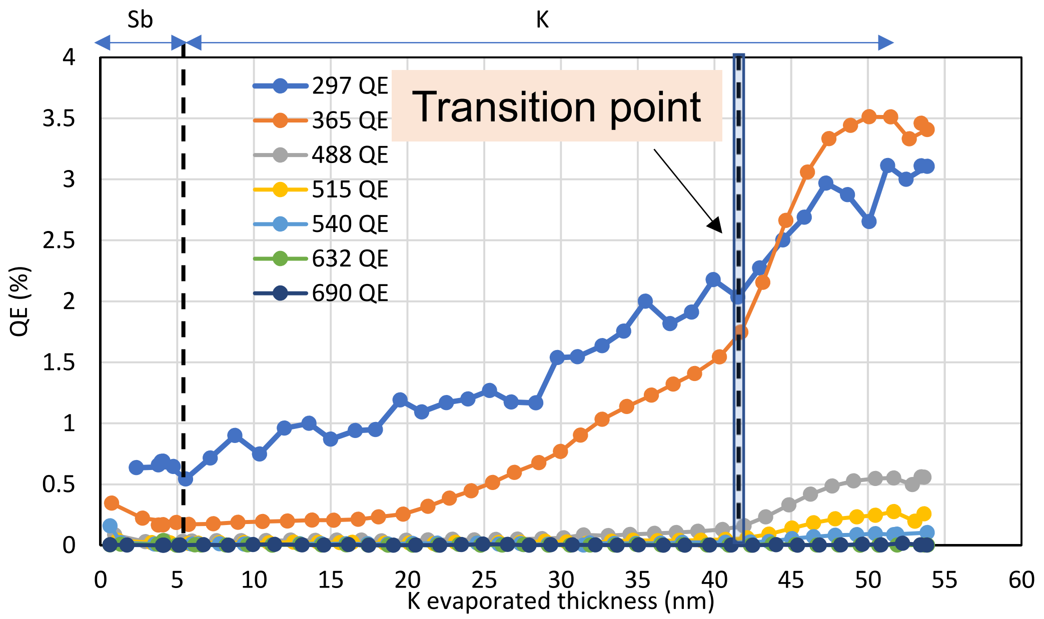


Decrease the power compared to the previous K deposition (137.2)

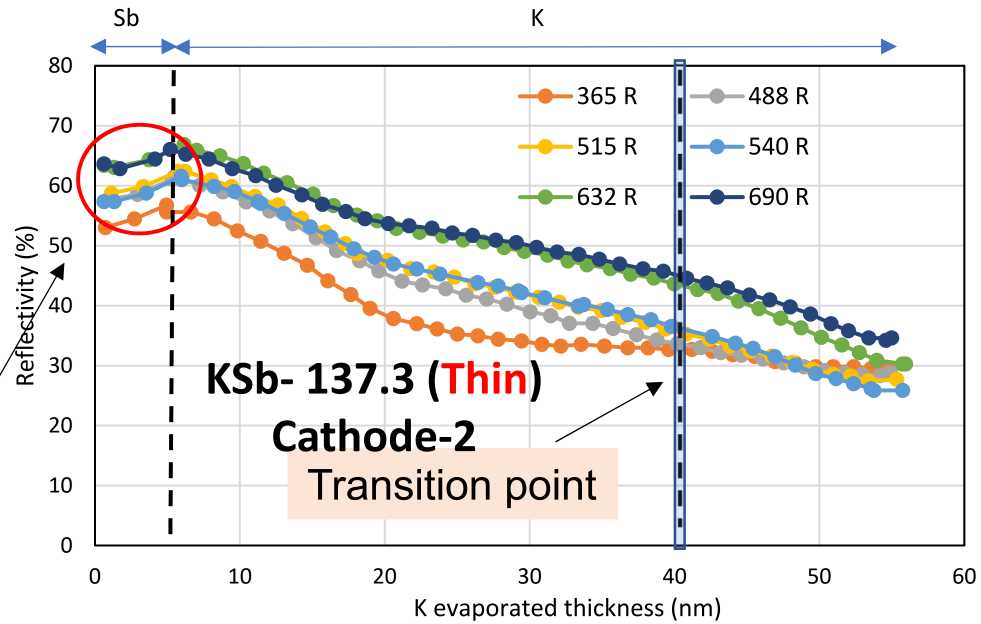
Decrease the power compared to the previous Cs deposition (137.2)

“Real-time” QE & Reflectivity curve 137.3 (KSb – Thin, Cathode-2)

- “Real-time” **QE** vs. evaporated **thickness** during Sb & K deposition
- “Real-time” **Reflectivity** vs. evaporated **thickness** during Sb & K deposition

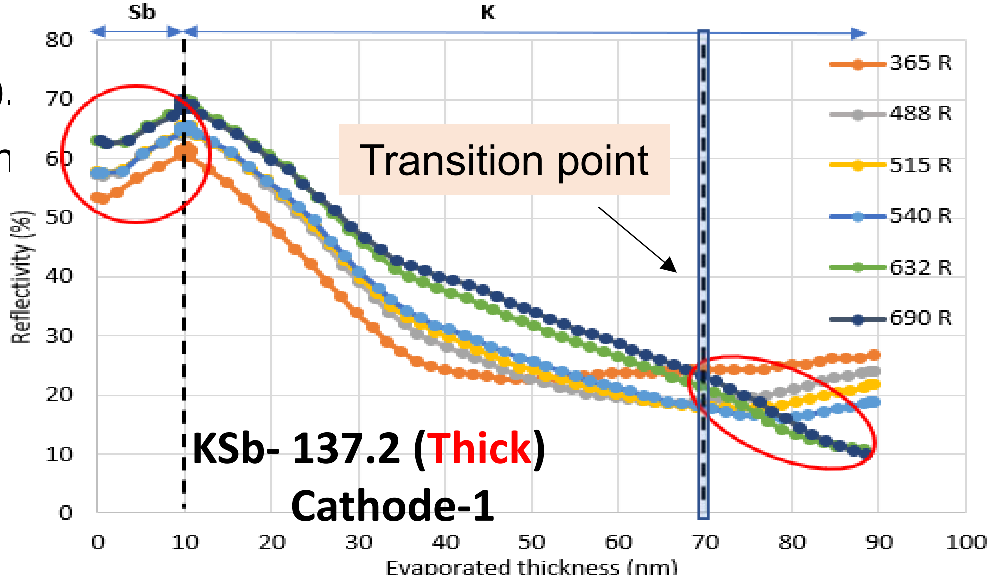


Transition point



KSb- 137.3 (Thin)
Cathode-2

Transition point



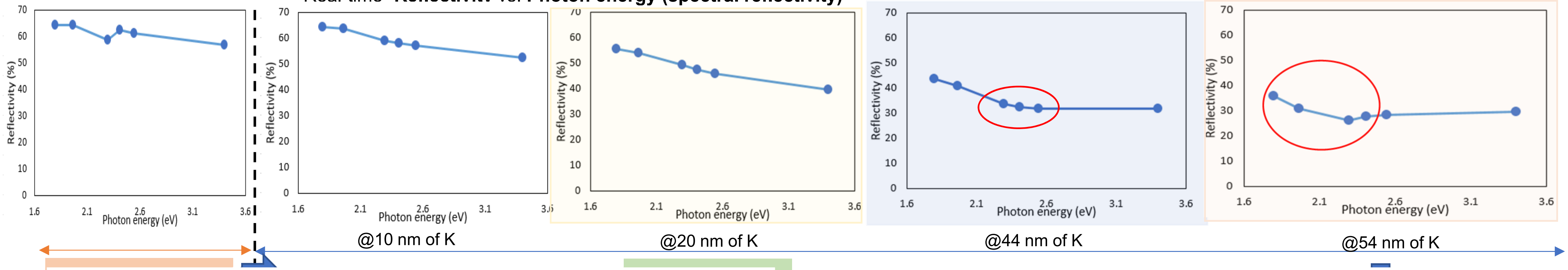
Transition point

KSb- 137.2 (Thick)
Cathode-1

- During **Sb** deposition, the **reflectivity** was **increased** (at **1.5** nm).
- There is a **change in slope** in real-time **QE** (at all the wavelength een observed after **41 nm** of K evaporation (transition point).
- The rate of QE increase at 365 was higher compared to 297 nm, (e-e scattering, if hν ≥ 2Eg).

Real-time Reflectivity history during Sb+K deposition (137.3 KSb-Thin, Cathode-2)

- “Real-time” **Reflectivitv** vs. **Photon energy (spectral reflectivity)**



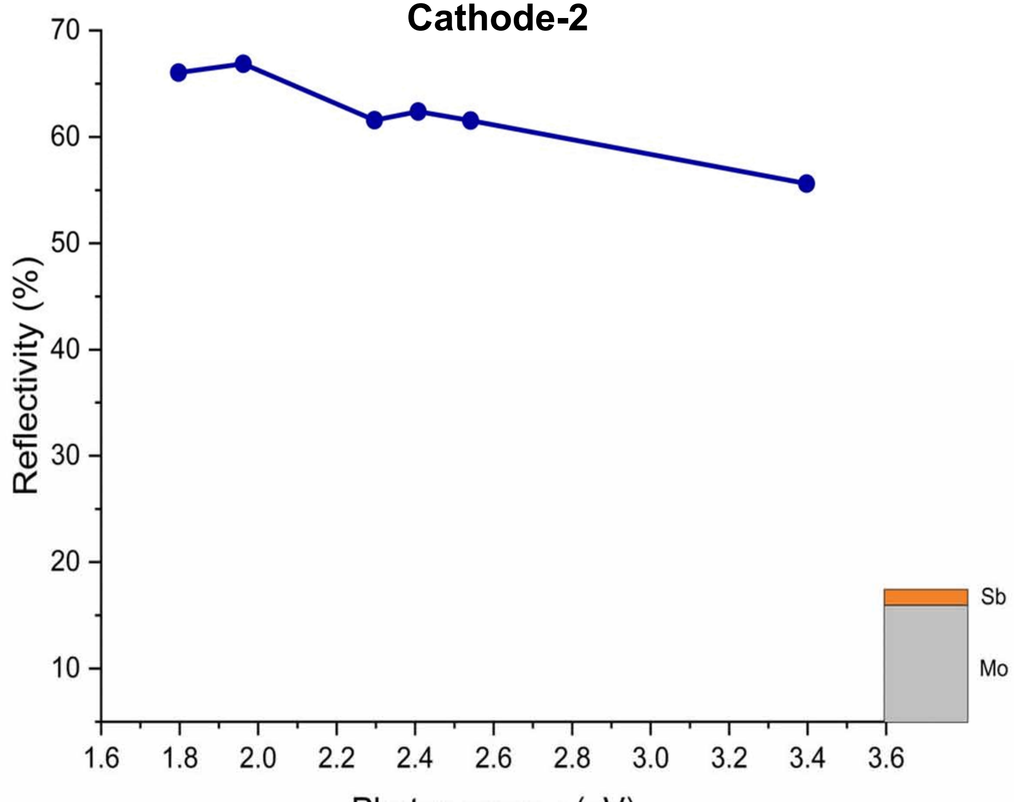
@10 nm of K

@20 nm of K

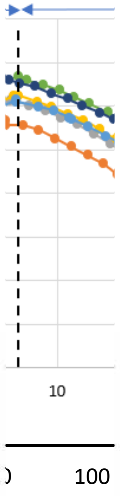
@44 nm of K

@54 nm of K

R(%) history during Sb+K deposition (137.3 KSb-Thin) Cathode-2



“Real



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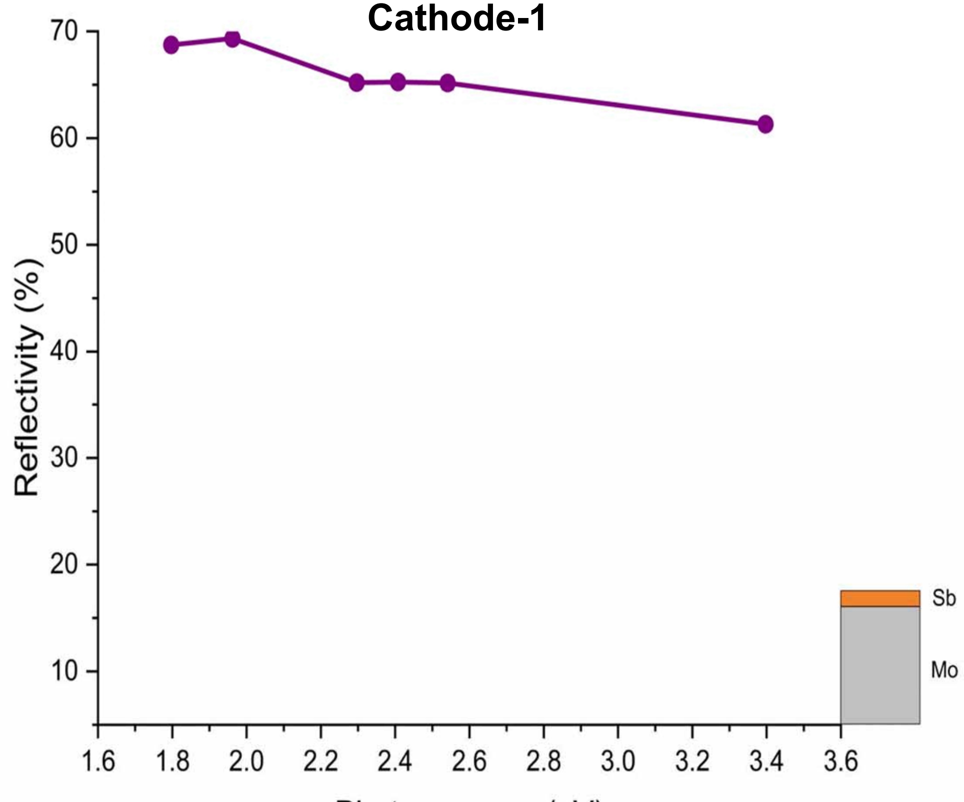
d at all

change

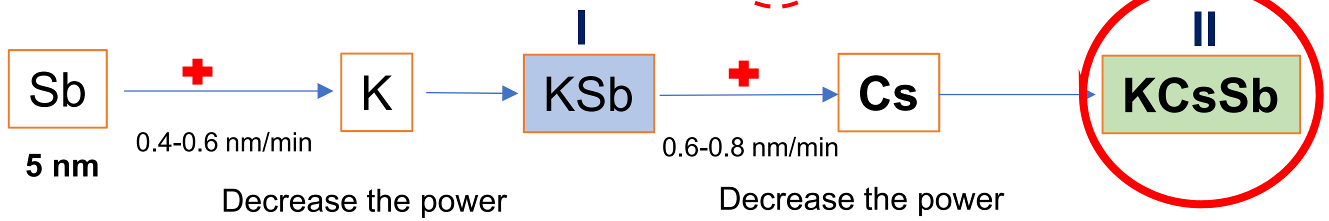
R(%) history during Sb+K deposition (137.2 KSb-Thick) Cathode-1



ture).

Cathode - 2

Cathode 137.3 (K+Sb+Cs), Sb = 5 nm (Thin)



5 nm

0.4-0.6 nm/min

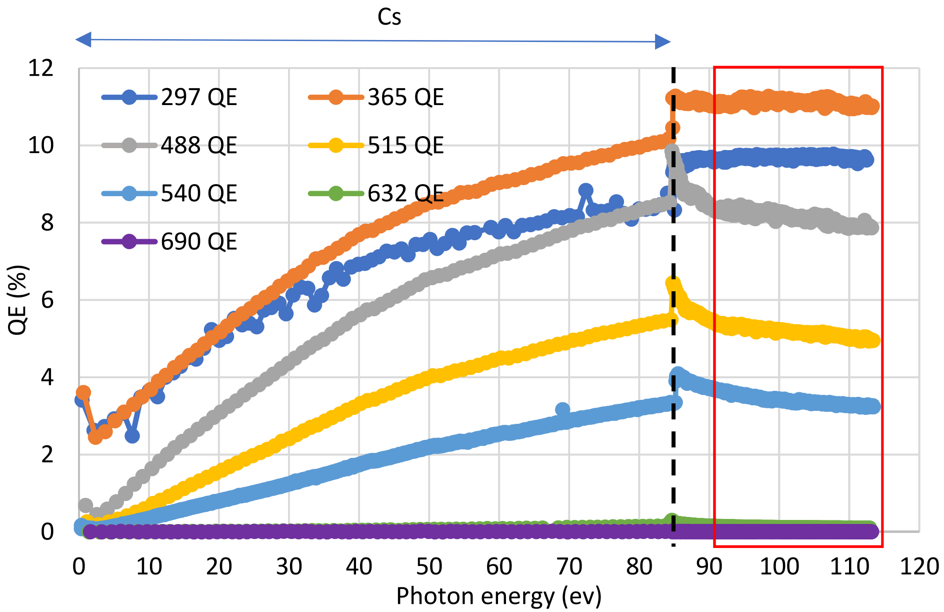
Decrease the power compared to the previous K deposition (137.2)

0.6-0.8 nm/min

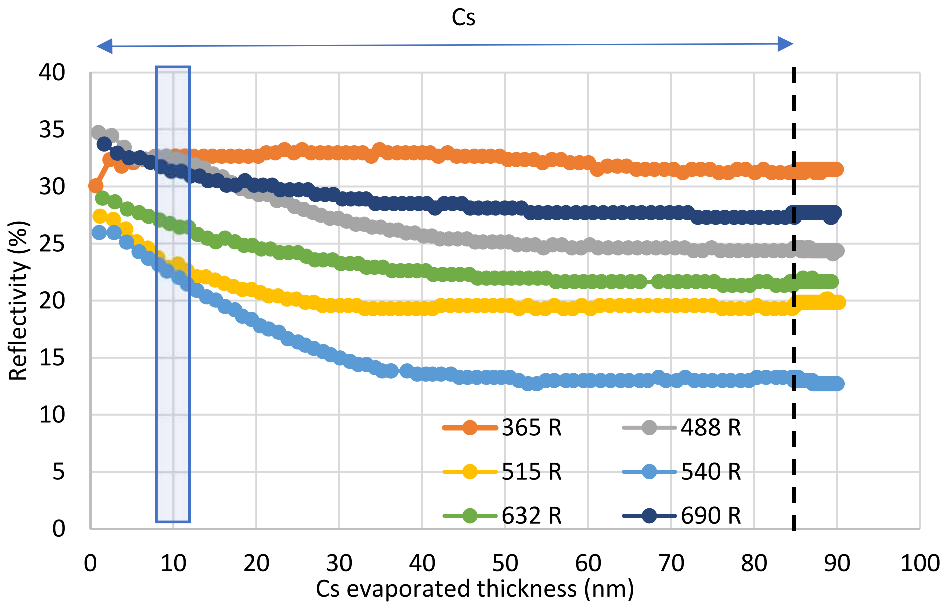
Decrease the power compared to the previous Cs deposition (137.2)

“Real-time” QE & Reflectivity curve 137.3 (KSb+Cs – Thin, Cathode-2)

- “Real-time” **QE** vs. evaporated **thickness** during Cs deposition



- “Real-time” **Reflectivity** vs. evaporated **thickness** during Cs deposition



- After **10 nm** of Cs evaporation, there is a **change in slope** in the **reflectivity** curve for **all the wavelengths** (except 365 nm).
- At the end of Cs evaporation, a small jump appeared at the QE (at all the wavelengths), but afterward, the QE was getting decreased except UV wavelengths (297 & 365 nm) (temperature-sensitive surface layer! [1]).

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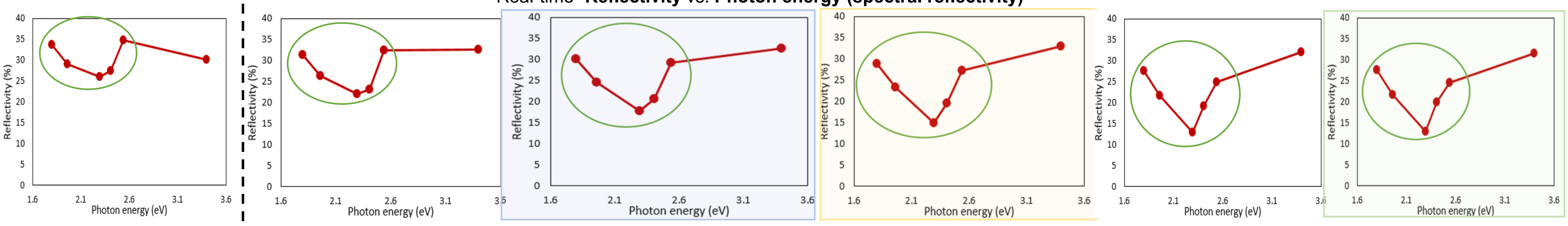
[1] Mohanty SK, Krasilnikov M, Oppelt A, Stephan F, Sertore D, Monaco L, Pagani C, Hillert W. Development and Characterization of Multi-Alkali Antimonide Photocathodes for High-Brightness RF Photoinjectors. Micromachines. 2023; 14(6):1182. https://doi.org/10.3390/mi14061182 .

Real-time Reflectivity history during KSb+Cs deposition (137.3 KCsSb-Thin, Cathode-2)

- “Real-time” **Reflectivity** vs. **Photon energy (spectral reflectivity)**



Sb

@10 nm of Cs

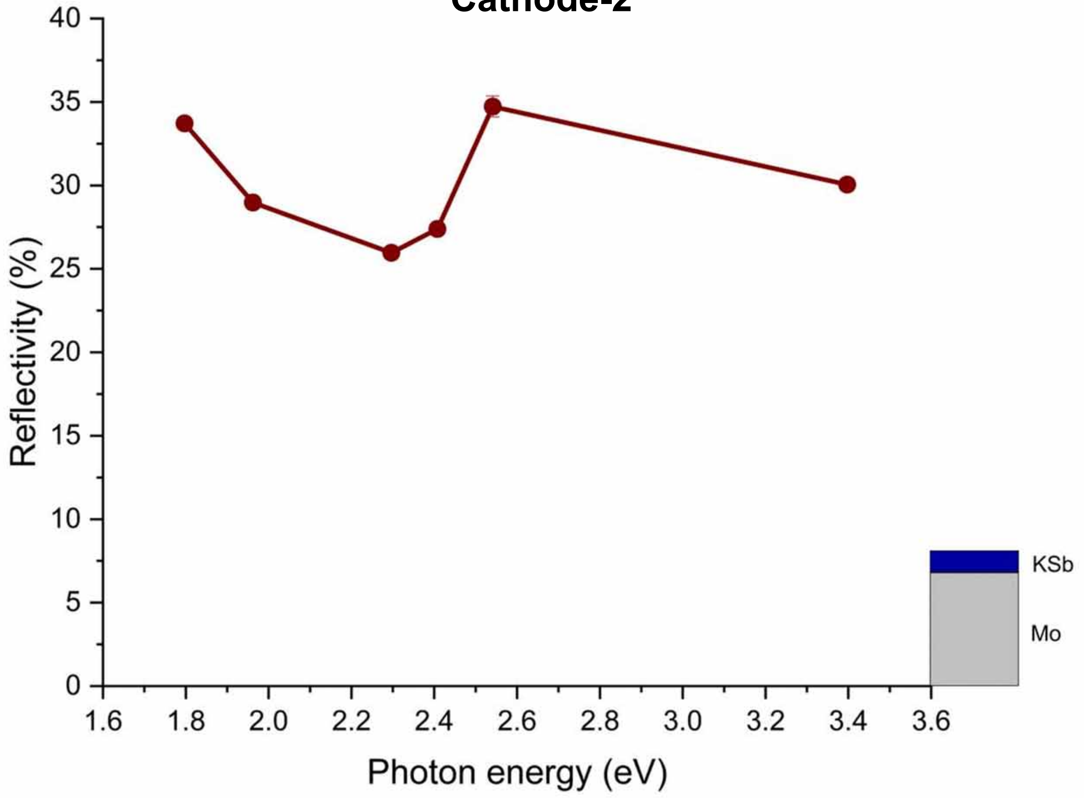
@20 nm of Cs

@30 nm of Cs

@60 nm of Cs

@85 nm of Cs

R(%) history during KSb+Cs deposition (137.3 KCsSb-Thin) Cathode-2



Rea

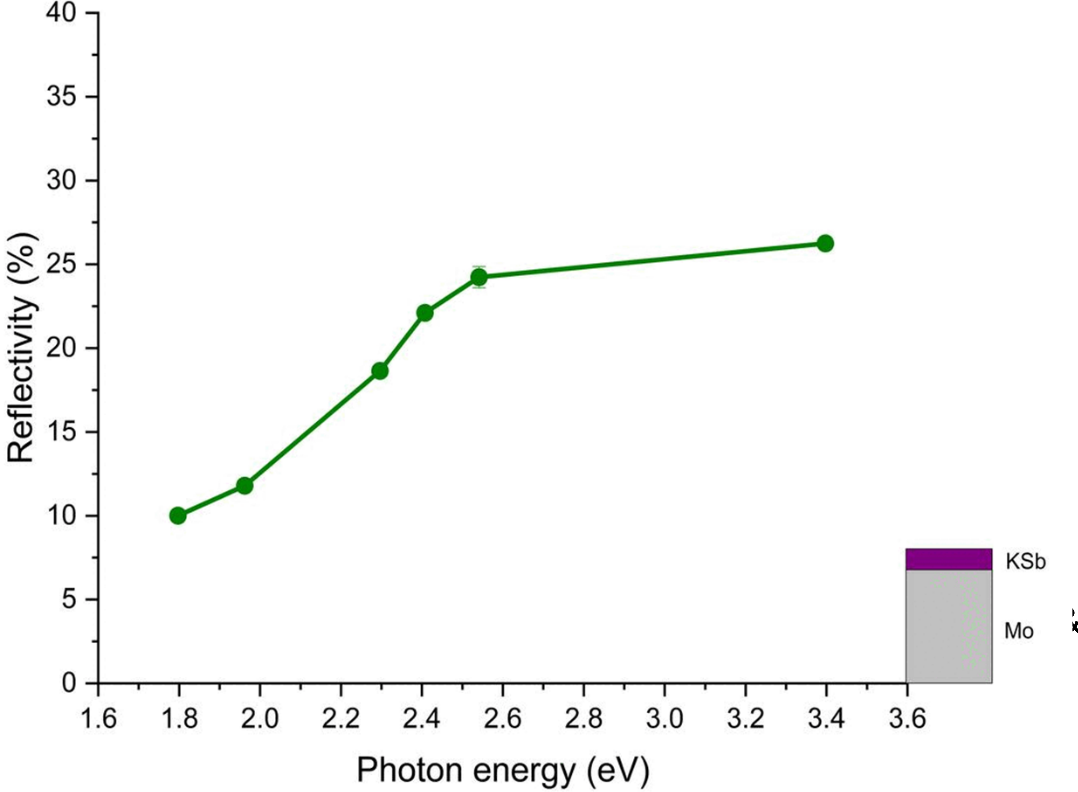
nick

rat

the

rat

R(%) history during KSb+Cs deposition (137.2 KCsSb-Thick) Cathode-1



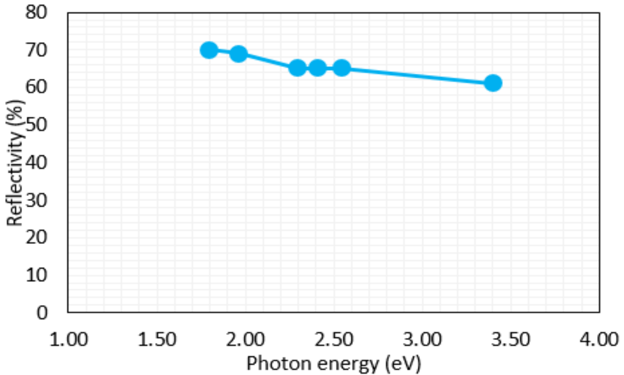
- F
  5
- U
- U

Comparison of R (%) between KCsSb **Thick** (137.2) and **Thin** (137.3) cathodes

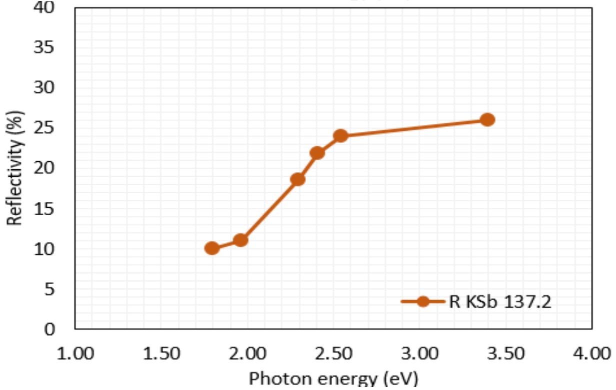
KCsSb **Thick** (137.2)

Spectral Reflectivity

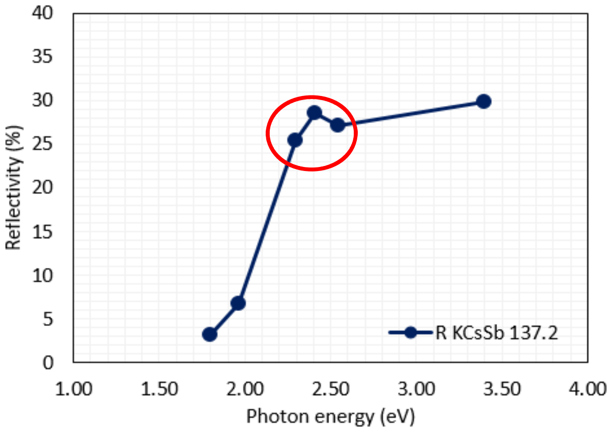
After **Sb**



After **K**



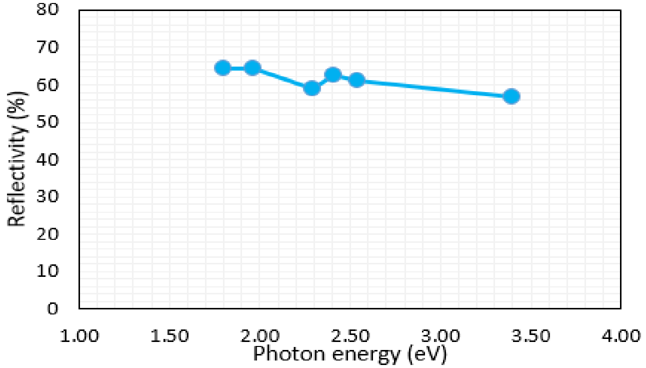
After **Cs**



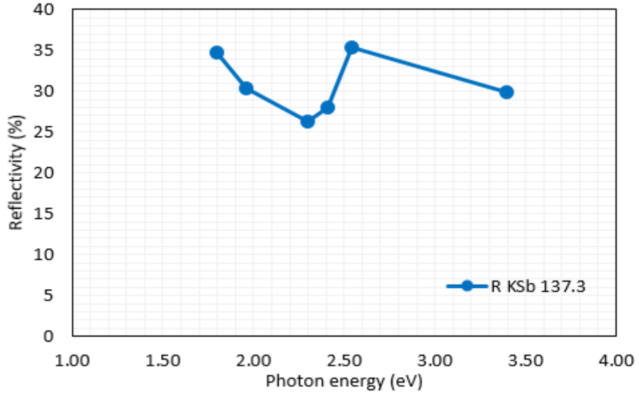
KCsSb **Thin** (137.3)

Spectral Reflectivity

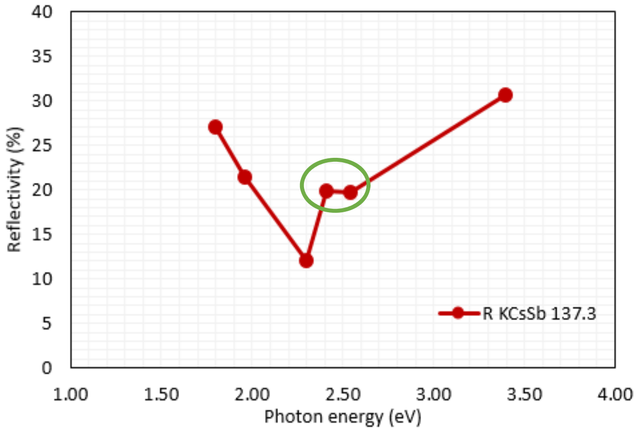
After **Sb**



After **K**



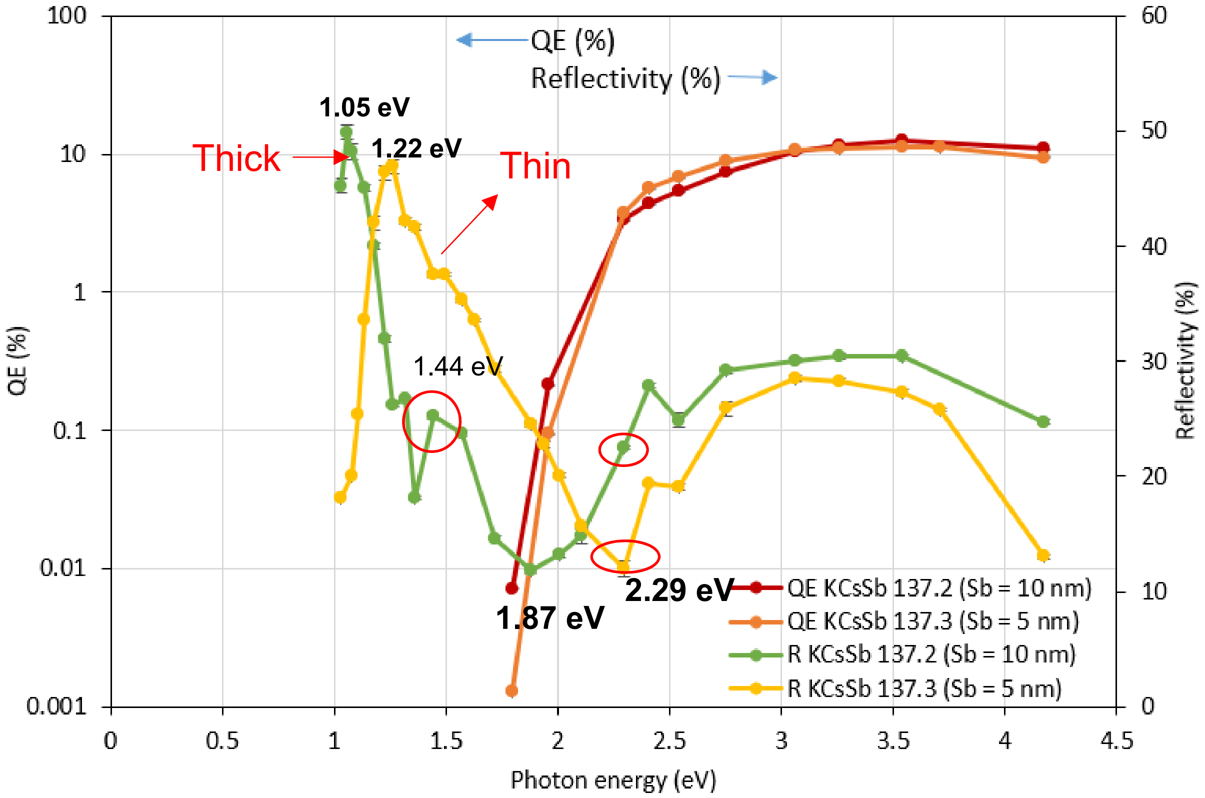
After **Cs**



Comparison between KCsSb Thick (137.2) and Thin (137.3) cathodes

KCsSb Thick (137.2)

KCsSb Thin (137.3)



- Colour of the cathode

- Colour of the cathode

Simulation results

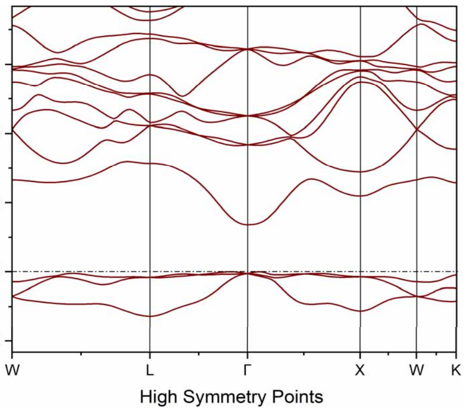
Density Function Theory (DFT) study

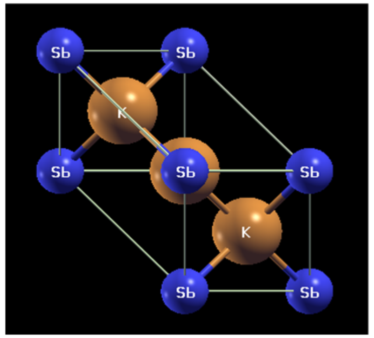
Electronic structure (DFT study)

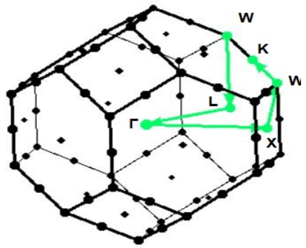
- Band structure calculation by **HSE method**

	K_3Sb (Cubic) direct gap	K_3Sb (Hexagonal) direct gap	K_2CsSb (Cubic) direct gap
Egap (K points)	**1.42 eV**	**1.05 eV**	**1.49 eV**
Experimental	1.4 eV [1]	1.1 eV [2]	1.2 eV [3]

K_3Sb (Cubic)

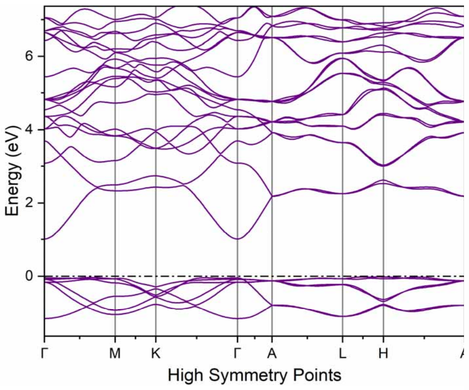


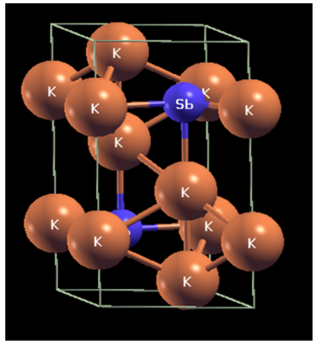


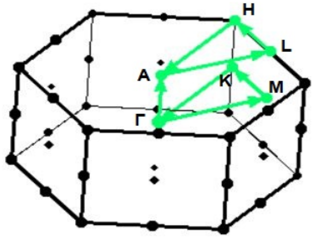


a = 8.493 angstrom

K_3Sb (Hexagonal)

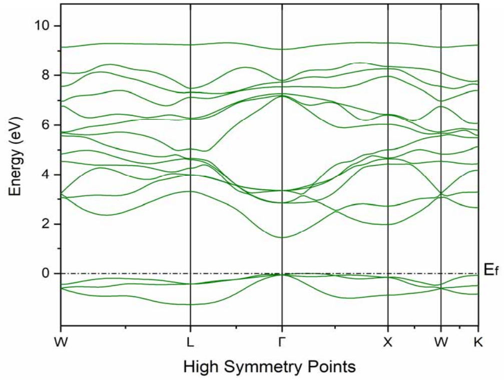


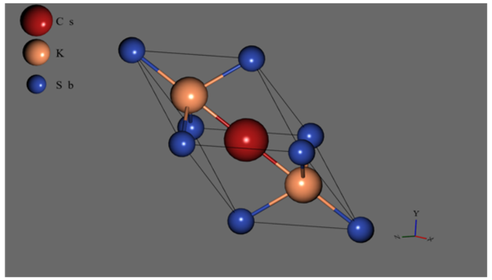


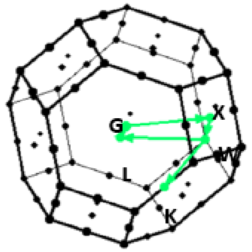


a = 6.025 angstrom
C = 10.690 angstrom

K_2CsSb (Cubic)







a = 8.7587 angstrom

[1] **A. H. Sommer** and W. H. McCarroll, “A New Modification of the Semiconducting Compound K3Sb,” Journal of Applied Physics, vol. 37, no. 1, pp. 174–179, 062004.
[2] A. H. Sommer and W. H. McCarroll, “A New Modification of the Semiconducting Compound K3Sb,” Journal of Applied Physics, vol. 37, no. 1, pp. 174–179, 06 2004.
[3] C. Ghosh and B. P. Varma, “Preparation and study of properties of a few alkali antimonide photocathodes,” Journal of Applied Physics, vol. 49, no. 8, pp. 4549–4553, 08 2008

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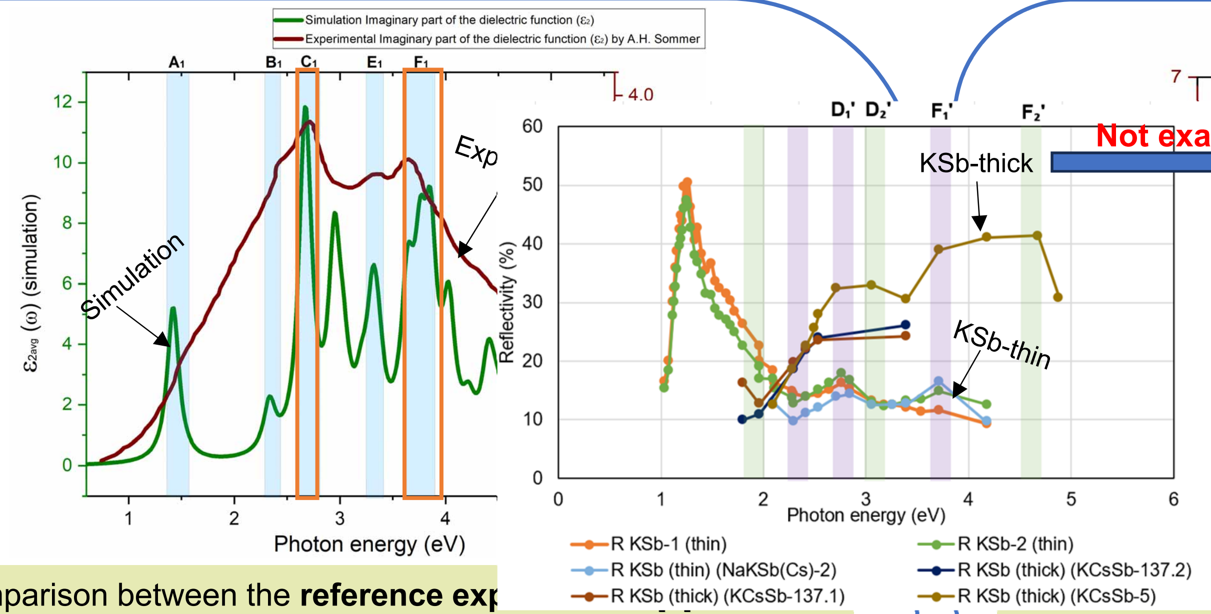
European XFEL

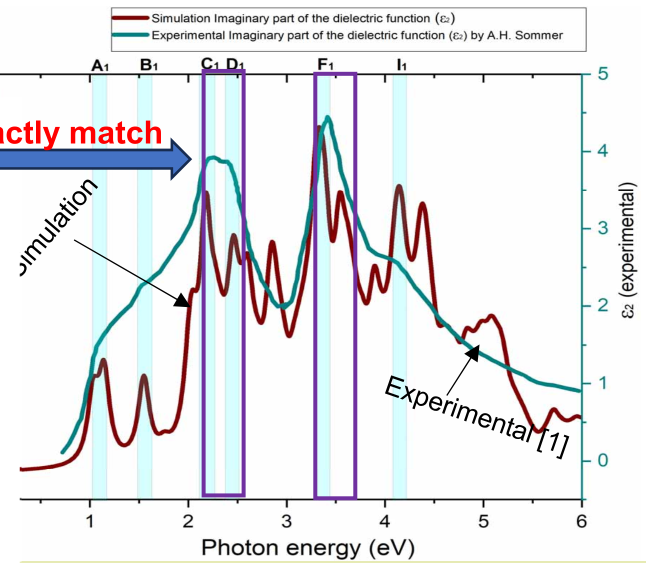
Comparison between the DFT Simulation and Experimental Data for K_3Sb

K_3Sb (Cubic)

K_3Sb (Hexagonal)



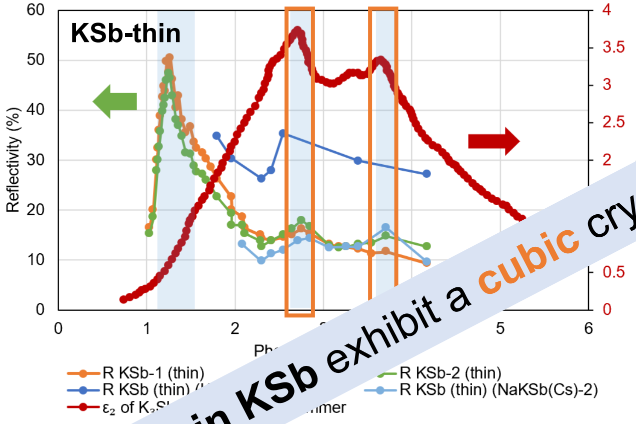
Not exactly match



Comparison between the **reference exp**

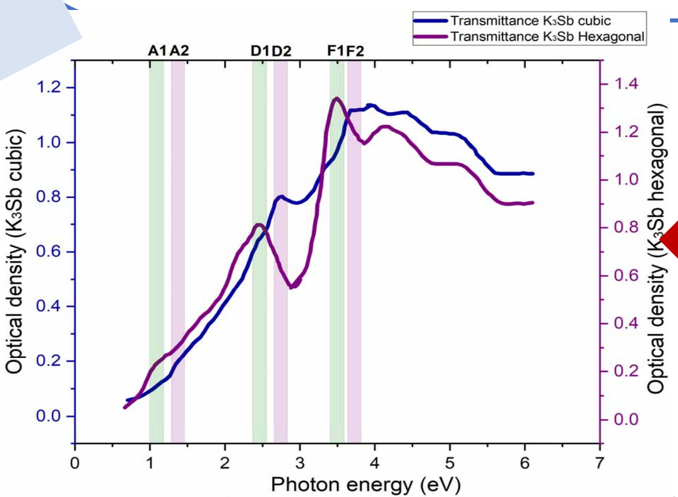
simulated **imaginary part of the dielectric function**

son between the **reference experimental** [1] and **simulated** **imaginary part of the dielectric function**



Potentially **thin KSb** exhibit a **cubic** crystal orientation!

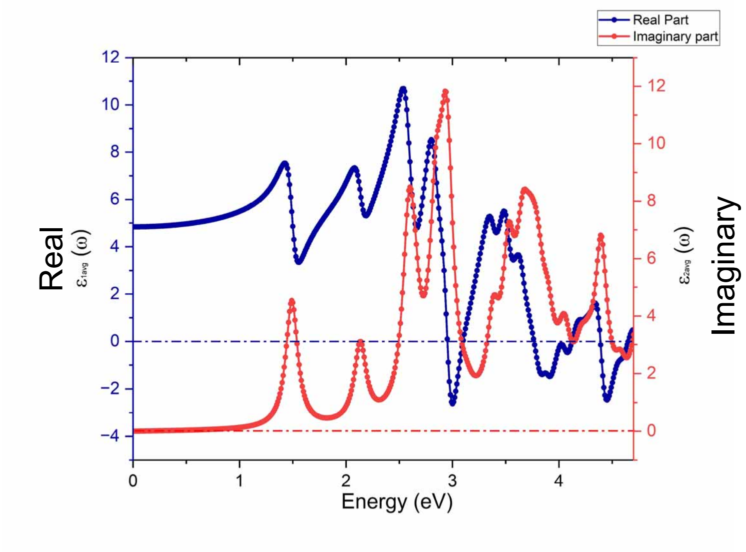
Comparison of t between reference experimental [1] and ab-grown **KSb (thin)** cathodes.



Comparing the **transmittance** data from reference experiments [1] for the **cubic** and **hexagonal** structures of **K_3Sb**.

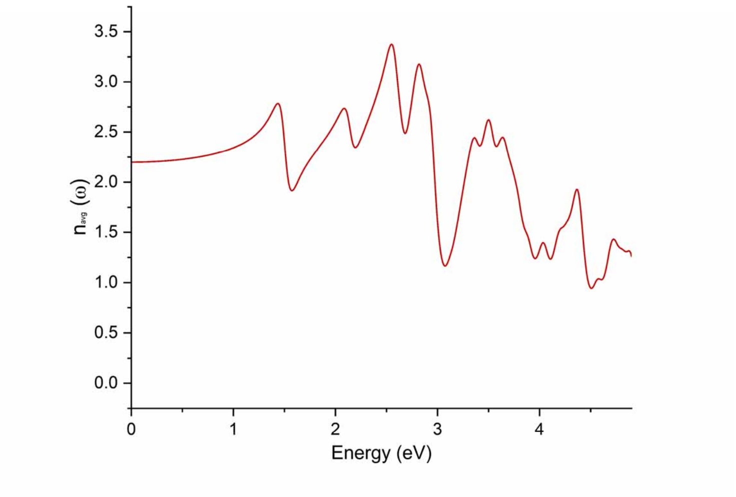
[1] **A. Ebina and T. Takahashi**, "Transmittance spectra and optical constants of alkali-antimony compounds k3sb, na3sb, and na2ksb," Phys. Rev. B, vol. 7, pp. 4712–4719, May 1973.

K$_2$CsSb (Cubic) Optical Properties

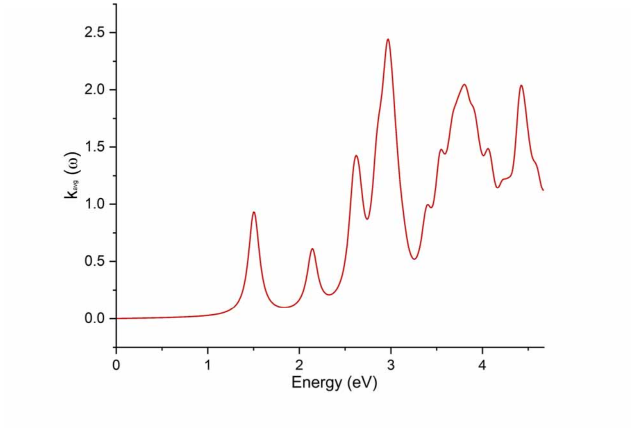


- Real & imaginary part of dielectric function

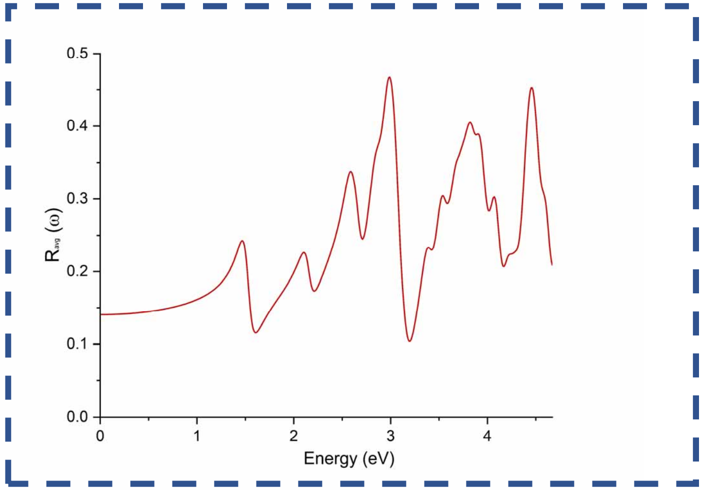
- $$n(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}-\varepsilon_1(\omega)}{2}\right]^{0.5}$$
- $$k(\omega) = \left[\frac{\sqrt{\varepsilon_1{}^2(\omega)+\varepsilon_2{}^2(\omega)}+\varepsilon_1(\omega)}{2}\right]^{0.5}$$
- $$R(\omega) = \frac{(1-n)^2+k^2}{(1+n)^2+k^2}$$
- $\alpha(\omega) = 4\pi k/\lambda$



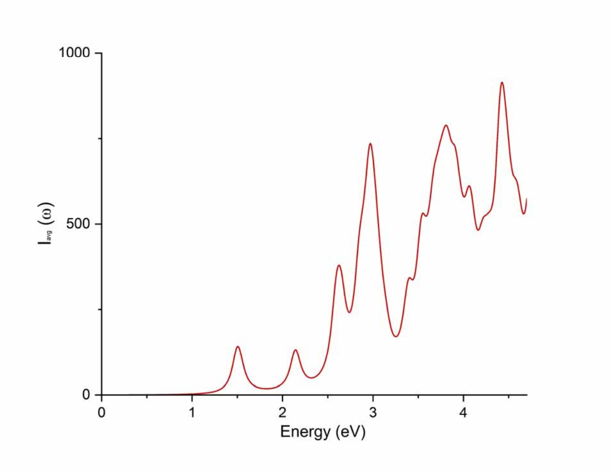
- Refractive index n(ω)



- Extinction coefficient k(ω)



- Reflectivity R(ω)

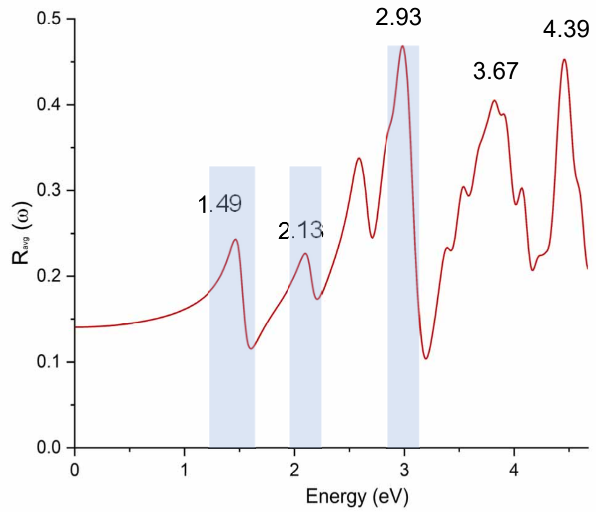


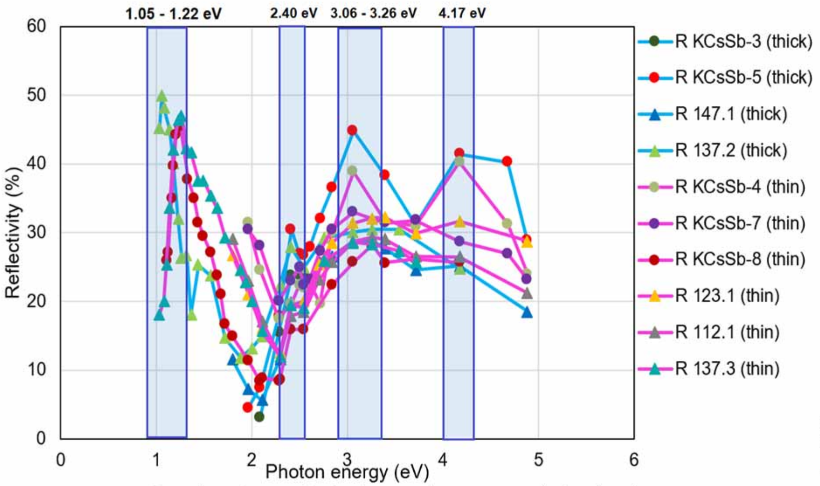
- Absorption coefficient I(ω)

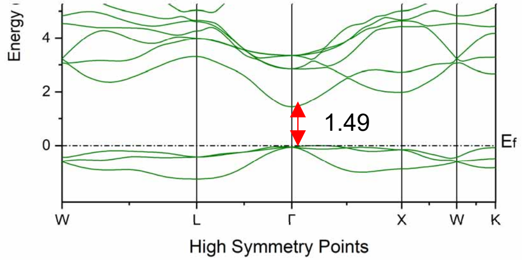
- Ground-state calculations, are implemented in Quantum Espresso and the dielectric function is calculated in epsilon.x post-processing utility.

Reflectivity comparison of KCsSb compound

- **Simulation Spectral Reflectivity K**

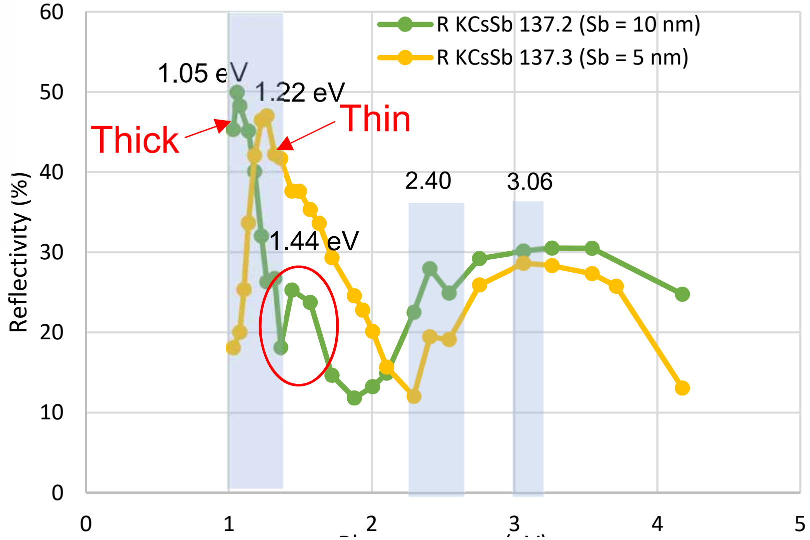


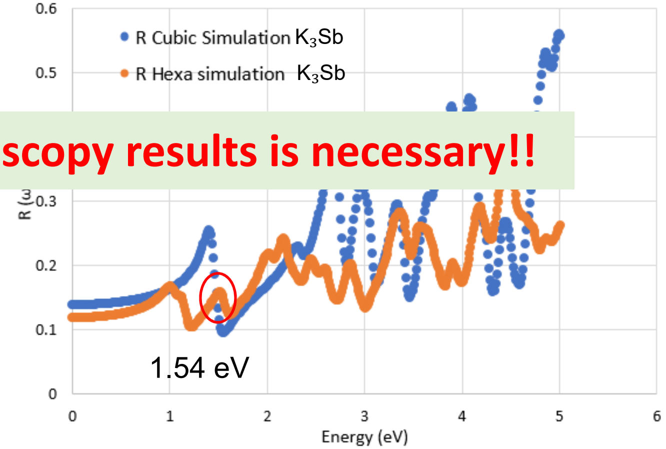




- Band structure

- **Experimental Spectral Reflectivity KCsSb**





To arrive at a definitive conclusion, verification with photoemission spectroscopy results is necessary!!

- Simulation Reflectivity **K_3Sb**

Summary & Future plan

- **Two cathodes** have been produced with sequential deposition with **QE** @514 nm recorded **4-6** %.
- A new **“multi-wavelength” Optical Diagnostics** setup has been used during the cathode deposition
  - ✓ It gives information about real-time spectral response and reflectivity during cathode growth.
  - ✓ The optical spectra of these semiconductors provide a rich source of information on their electronic properties.
- Comparing the **spectral reflectivity** between two cathodes shows that **the intermediate phase**, i.e., K+Sb (**KSb** compound), and **the final phase**, i.e., **KCsSb** compound, potentially contain **different crystal structures** for **thick** (Sb = 10nm) and **thin** (Sb = 5 nm) cathodes. (Further verification through photoemission spectroscopy results is required!)
- By **comparing with DFT simulation data**, it has been found that, potentially, **both the cathodes** (i.e., thin and thick) have a **different band gap**.
- **Analyzing these optical spectra**, especially spectral reflectivity, and **comparing** them with the theoretical model **(DFT results)** offers a **valuable method to predict the electronic structure** of the grown compound.

❑ Future Plans:

- The **next batch** of **green cathodes** is planned to be **tested at PITZ** at the end of this year.
- TRAnsverse Momentum Measurement (TRAMM) device is currently being developed and planned to be integrated into the production system to measure thermal emittance.
- Photoemission spectroscopy study.

Thank You for your attention!