

Ab Initio Many-Body Photoemission Model

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Motivation

- Understand photocathode materials, especially **alkali-antimonides**, with *ab initio* many-body methods

Previous works:

- Surface properties:

R. Schier et al., Phys. Rev. Mat. 6, 125001 (2022)

- High-throughput screening:

R. Schier et al., Adv. Theory Simul. 7, 2400680 (2024)

- Electronic band structure & optical spectrum:

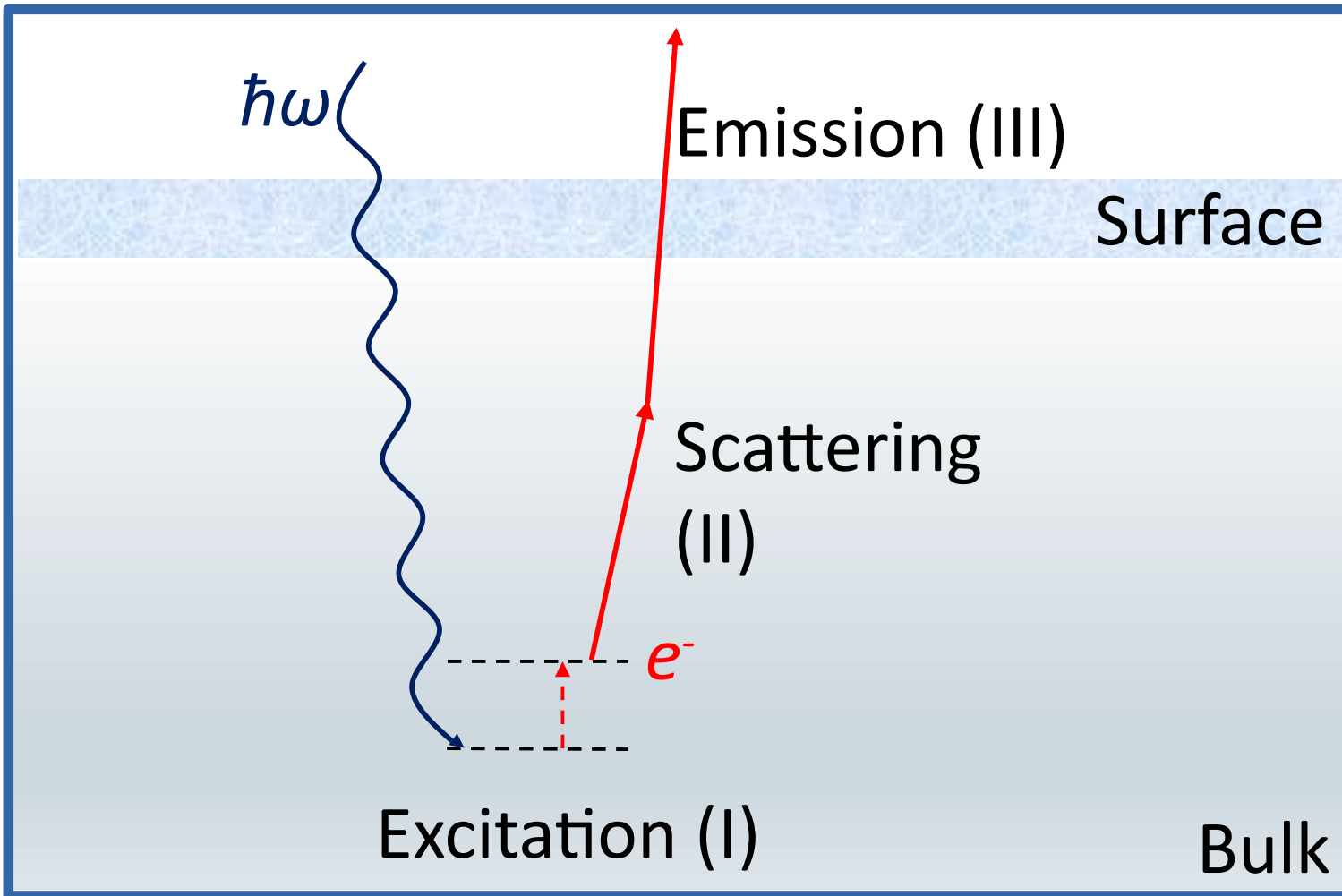
R. Schier & C. Cocchi, Phys. Rev. Mat. 9, 053804 (2025)

C. Xu et al., Electron. Struct. 7, 015001 (2025)



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Three-Step Model



Recent implementations based on density functional theory (DFT):

E. R. Antoniuk et al.,
Phys. Rev. B 101, 235447 (2020)

P. Saha et al.,
J. Appl. Phys. 133, 053102 (2023)

DFT is a ground-state theory –
no accurate band gap and
no electron-hole correlations!

**We present an implementation of
the three-step model with results
from many-body theory.**

W. E. Spicer, Phys. Rev. 112, 1 (1958)

C. N. Berglund & W. E. Spicer, Phys. Rev. 136, A1030 (1964)

Theory Workflow

DFT

Ground state: Kohn-Sham equations

$$\hat{h} |\phi_{nk}\rangle = \epsilon_{nk} |\phi_{nk}\rangle$$

W. Kohn and L. J. Sham, Phys. Rev. 140, A1133 (1965)



exciting

G_0W_0

Electron-electron correlation: Self-energy correction

$$\Sigma = iG_0W_0$$

→ Accurate band gap

$$\epsilon_{nk}^{QP} = \epsilon_{nk} + Z_{nk} [\text{Re}\Sigma_{nk}(\epsilon_{nk}) - V_{nk}^{xc}]$$

L. Hedin, Phys. Rev. 139, A796 (1965)

BSE

Electron-hole correlation: Bethe-Salpeter equation (BSE)

$$\hat{H}^{BSE} |A^\lambda\rangle = E^\lambda |A^\lambda\rangle$$

$$\hat{H}^{BSE} = \hat{H}^{diag} + 2\hat{H}^x + \hat{H}^c$$

Optical spectrum

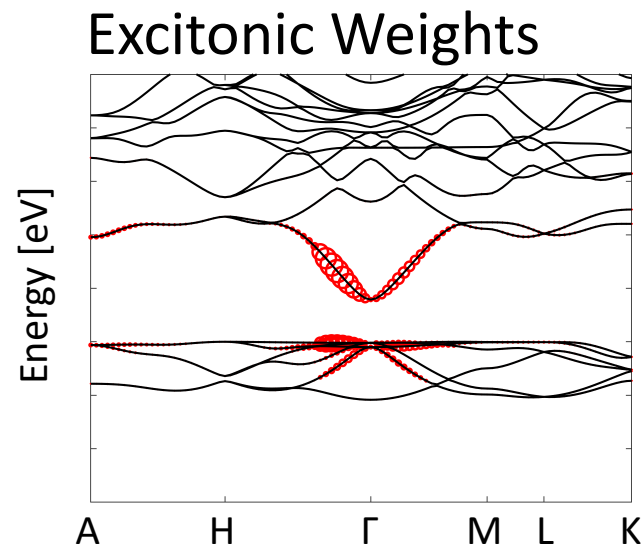
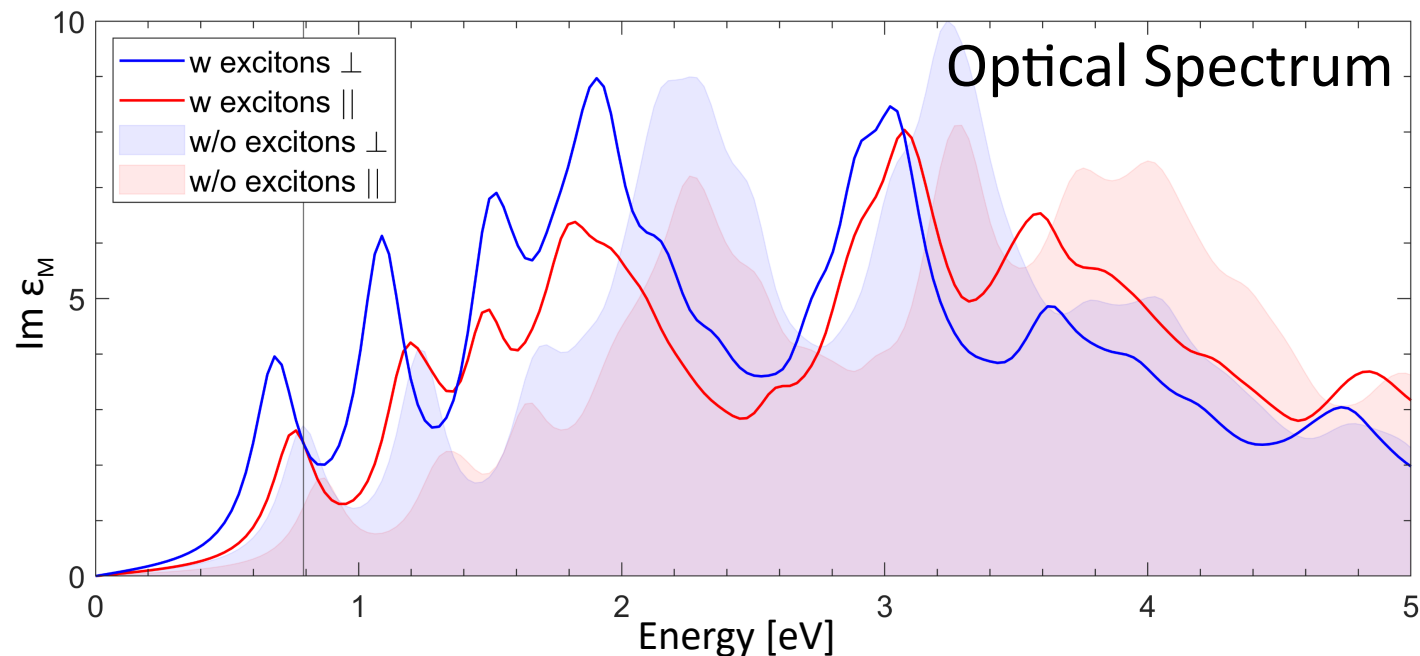
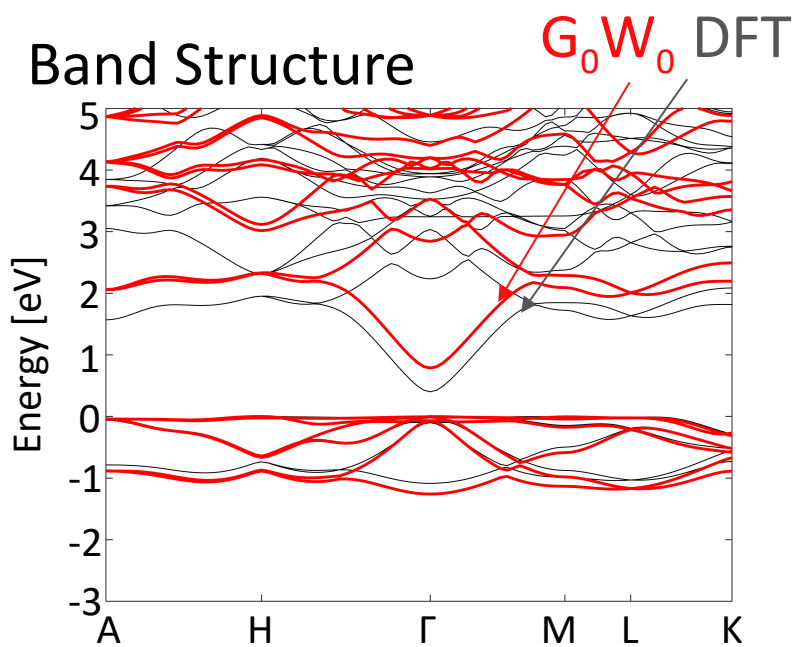
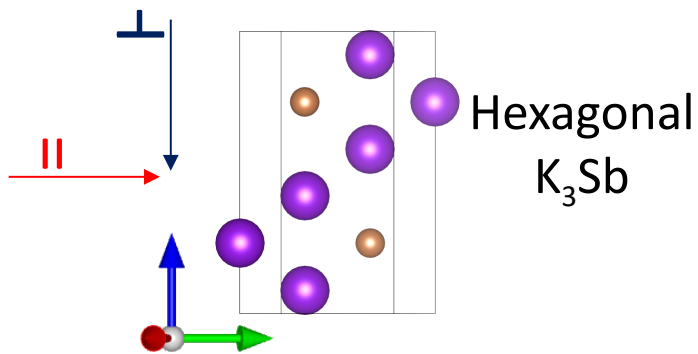
$$\text{Im } \epsilon_M = \frac{8\pi^2}{\Omega} \sum_{\lambda} |\mathbf{t}^\lambda|^2 \delta(\omega - E^\lambda)$$

Excitonic weights

$$w_{ck}^\lambda = \sum_v |A_{vck}^\lambda|^2$$

E. E. Salpeter and H. A. Bethe, Phys. Rev. 84, 1232 (1951)

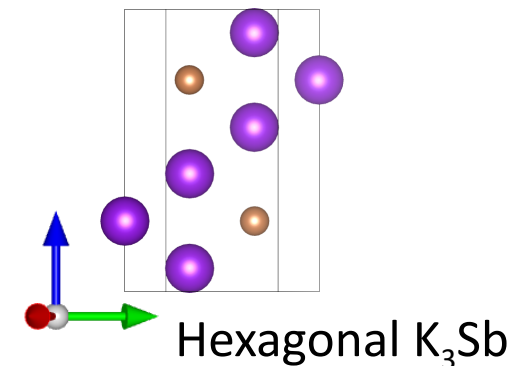
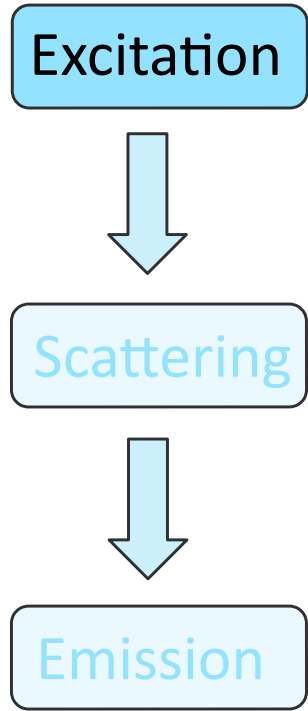
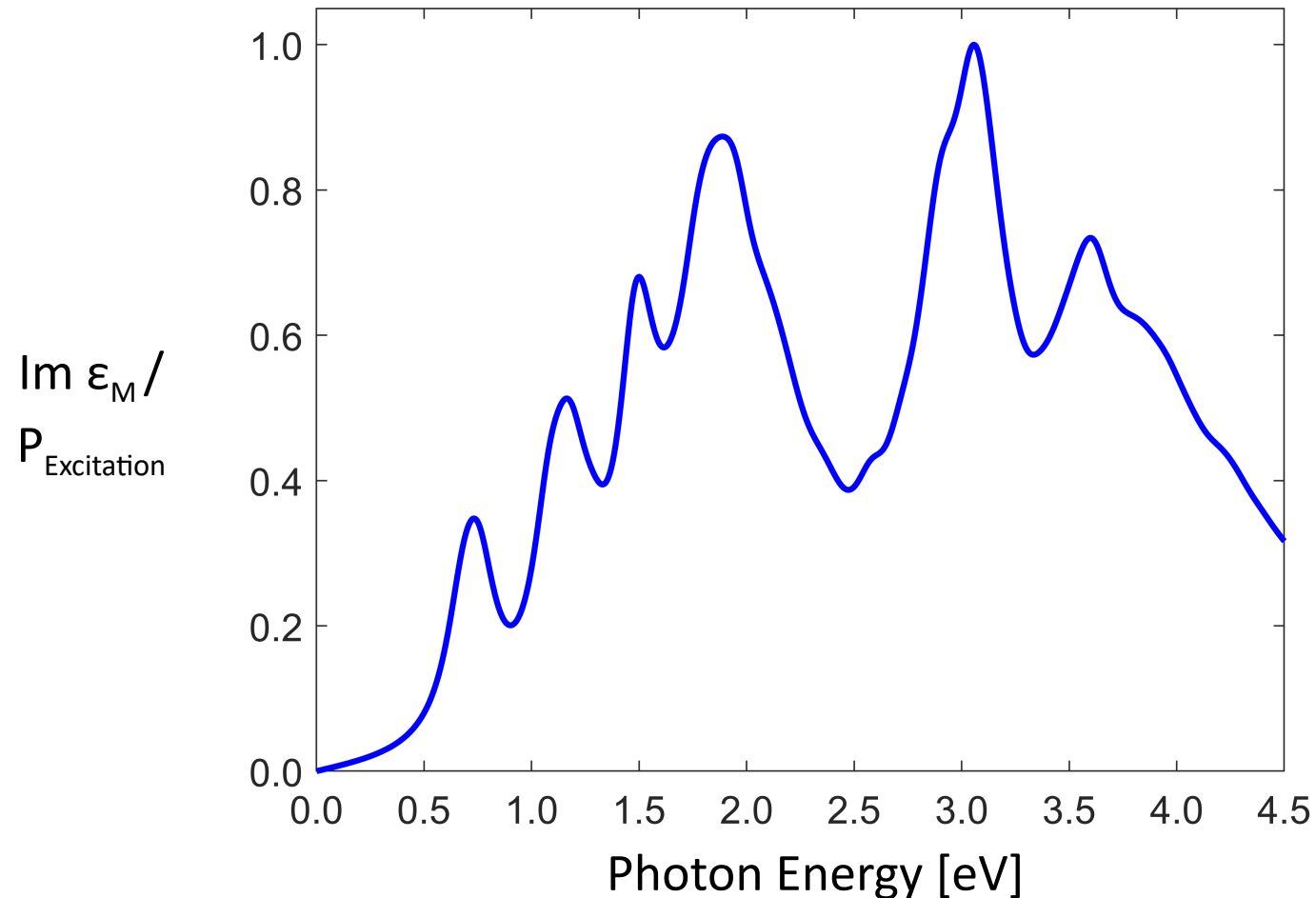
Electronic & Optical Properties of K_3Sb



R. Schier & C. Cocchi,
Phys. Rev. Materials 9, 053804 (2025)

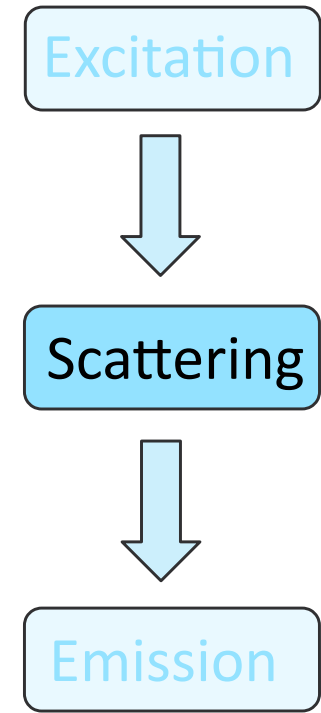
Step I: Excitation

$\text{Im } \epsilon_M$ averaged over all components
+ normalized to the highest peak



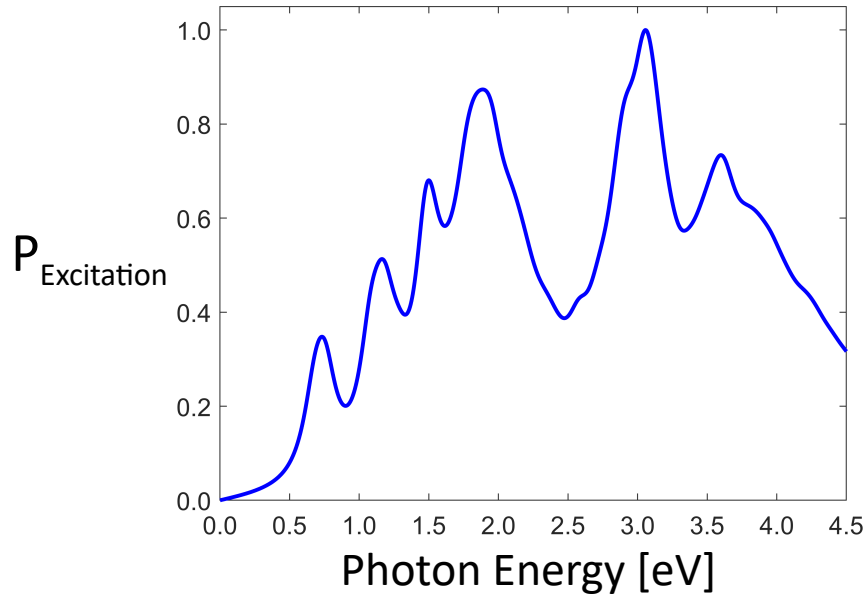
Step II: Transport / Scattering

- Scattering can be covered through Monte Carlo simulations.
[e.g. P. Gupta et al., J. Appl. Phys. 121, 215702 (2017)]
- However, inelastic scattering is small in semiconducting thin films.
- Only elastic scattering in this model.



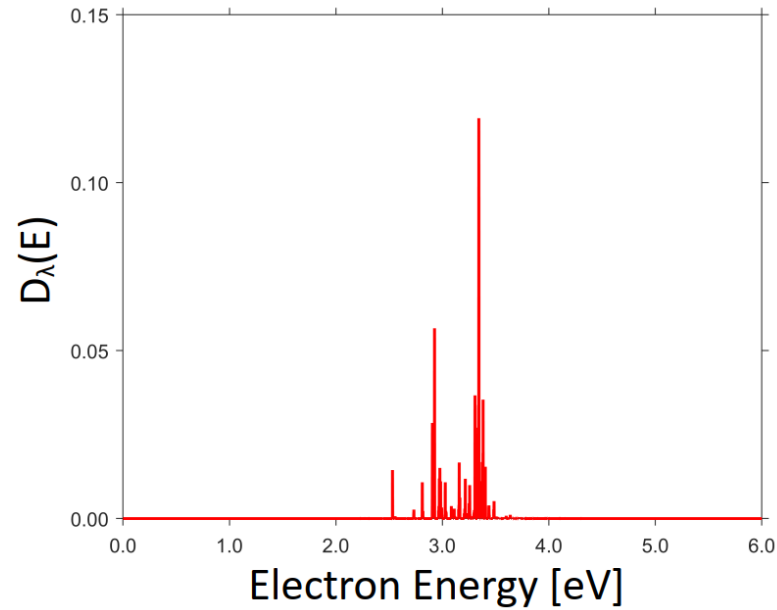
Step III: Emission

Optical Spectrum



+

Energy distribution of excited electrons



Excitation



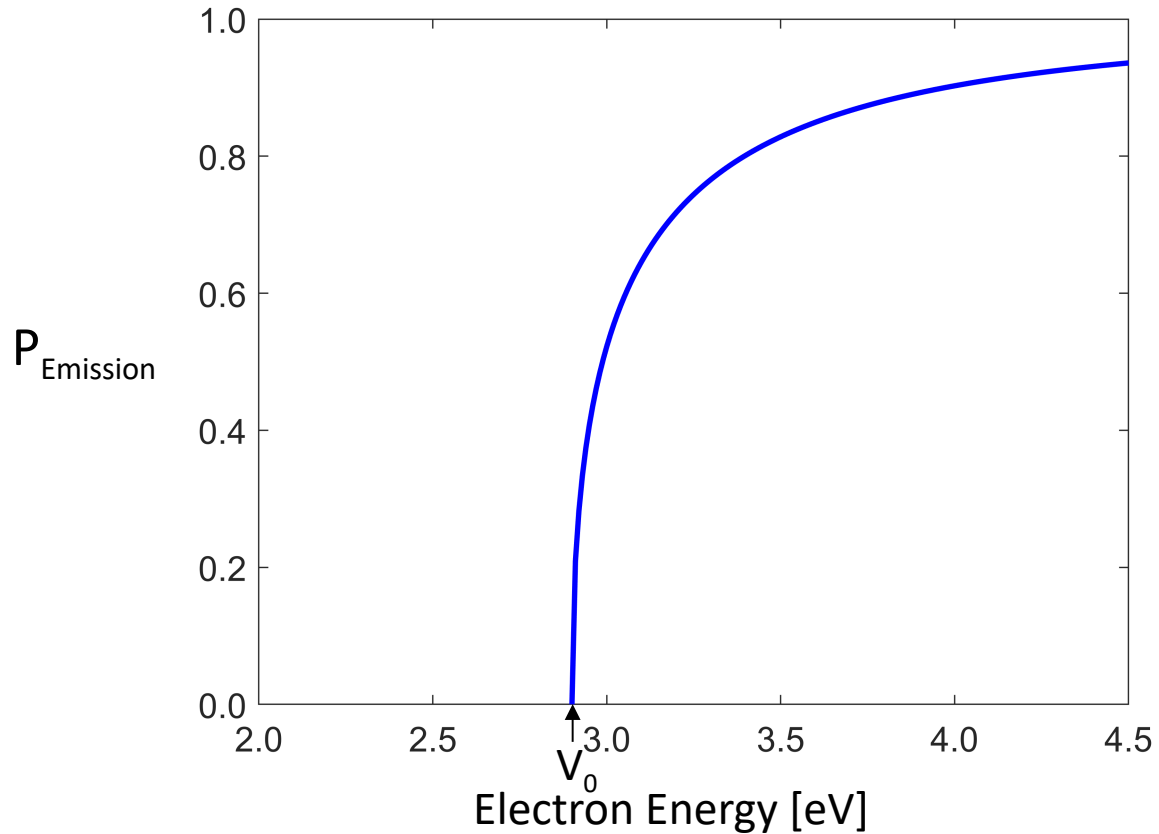
Scattering



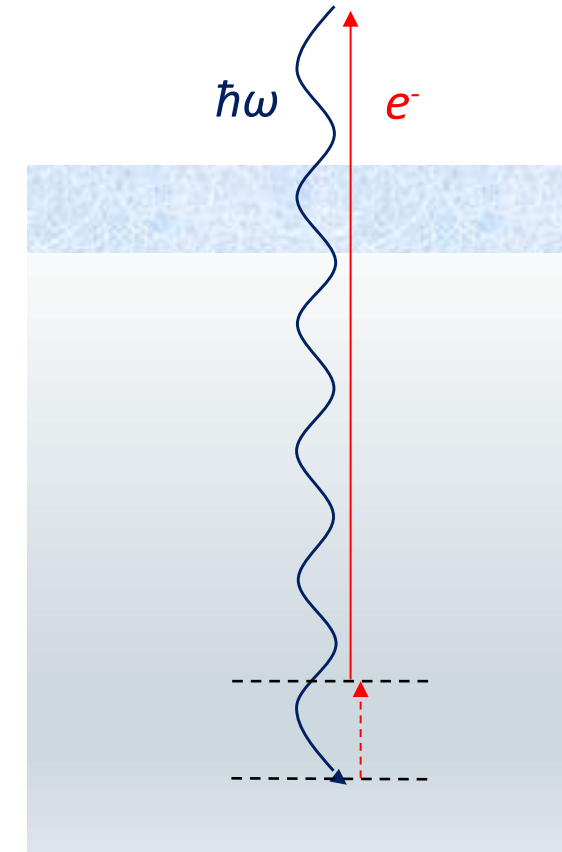
Emission

Emission Probability

- Simple approximation for $P_{\text{Emission}}(E)$: potential step



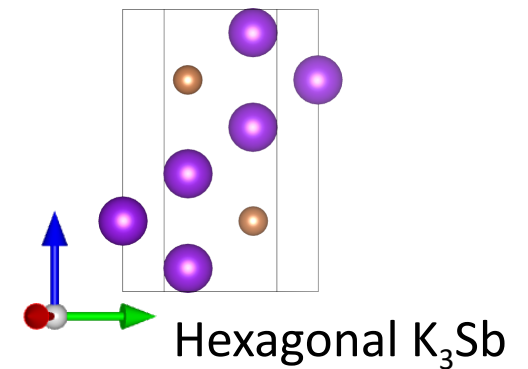
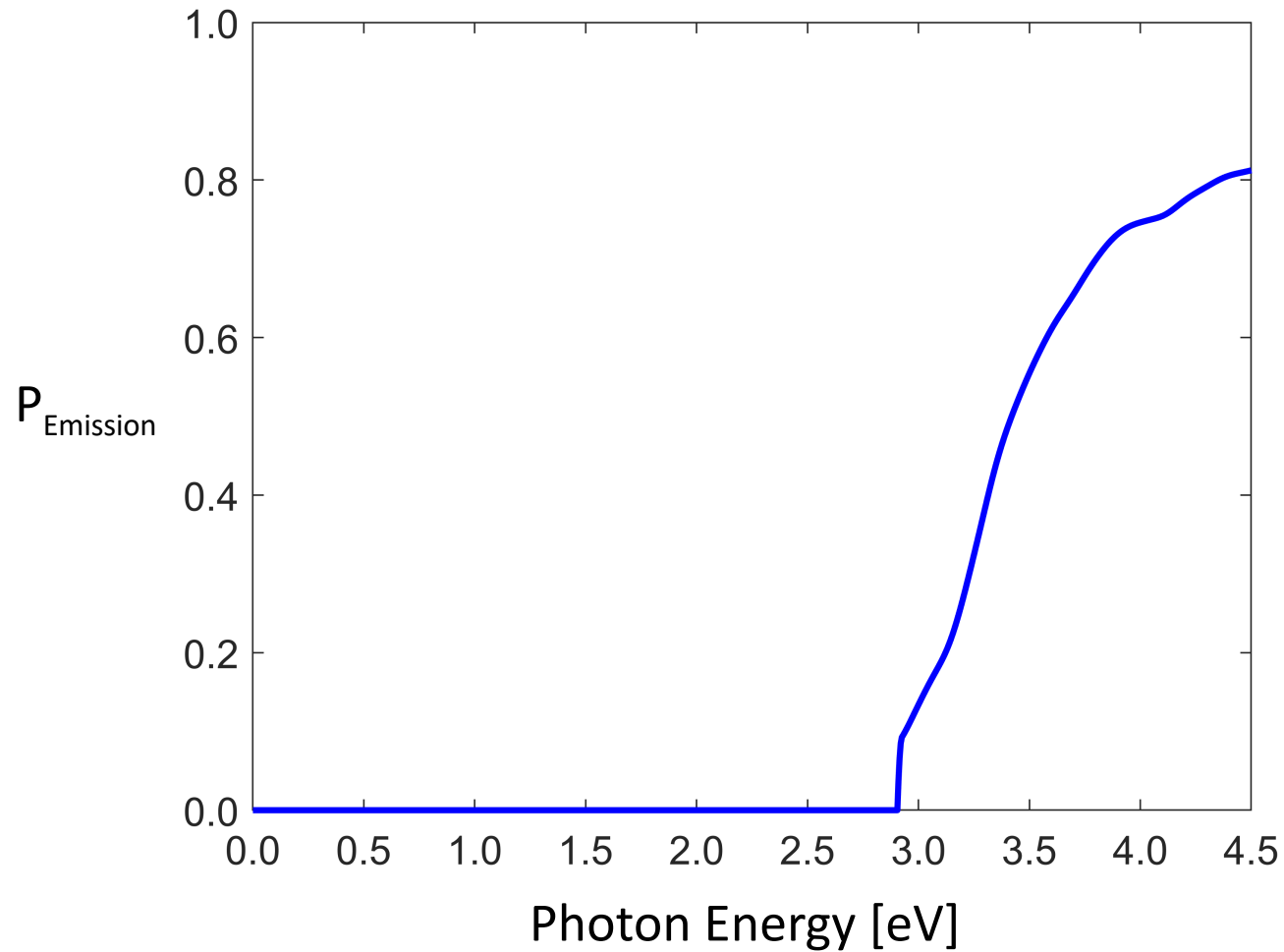
Here: V_0 from experiment
[W. E. Spicer, Phys. Rev. 112, 1 (1958)]



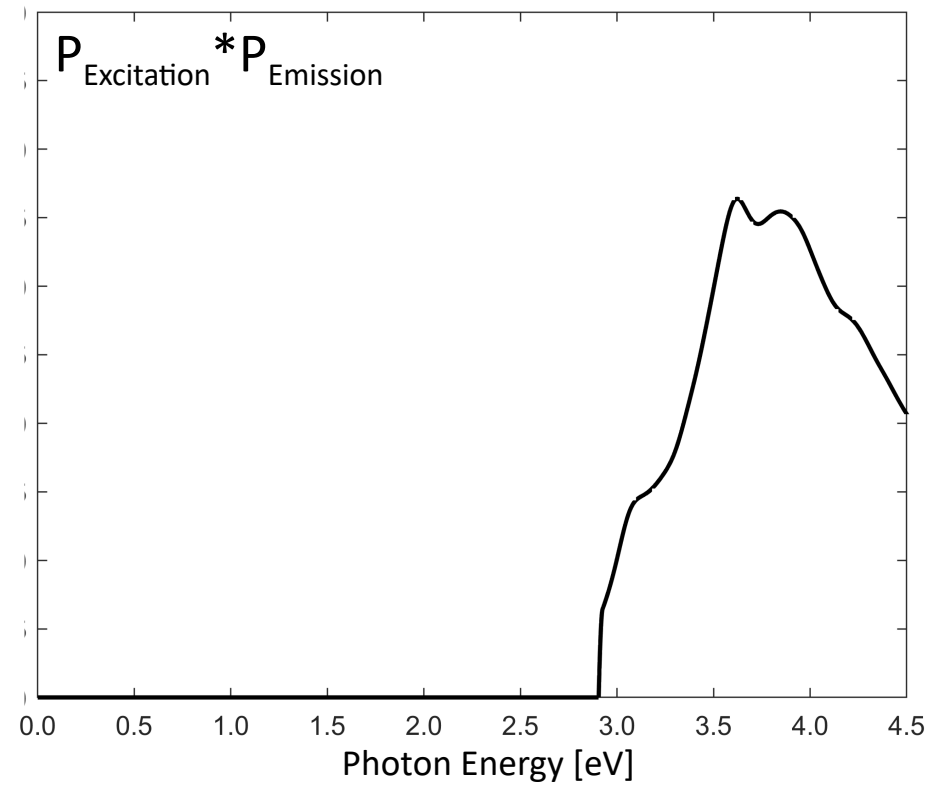
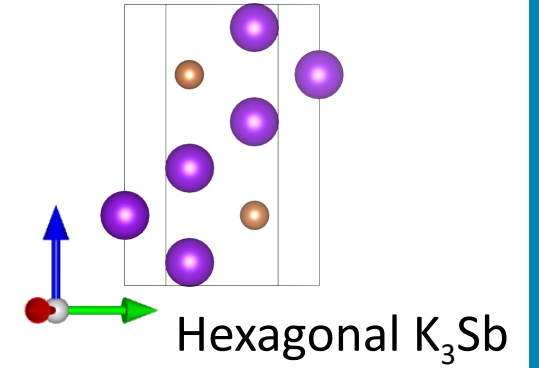
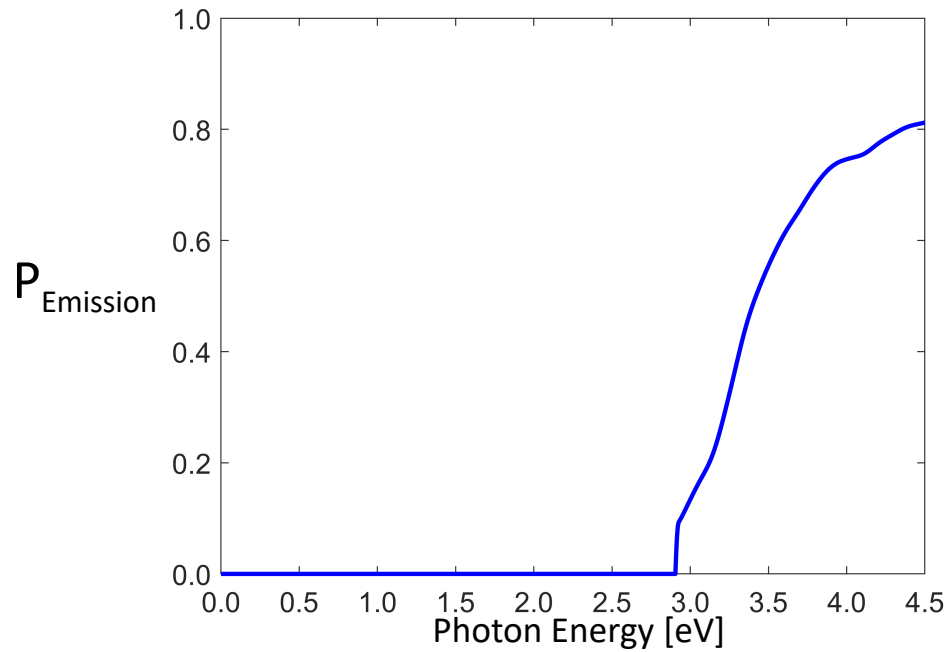
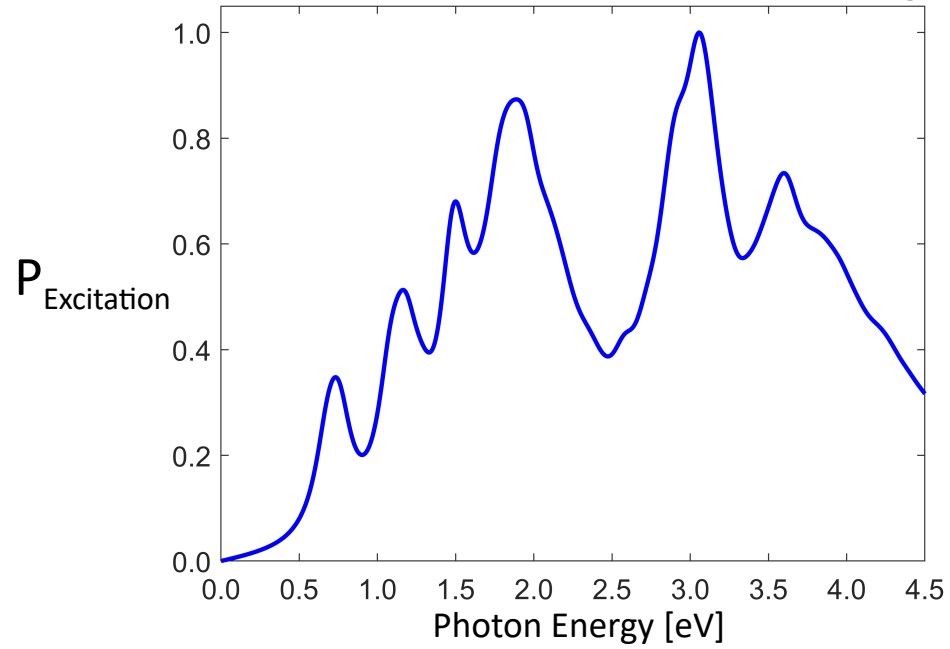
Alternative: surface slab calculation:

R. Schier et al., Phys. Rev. Mat. 6, 125001 (2022)

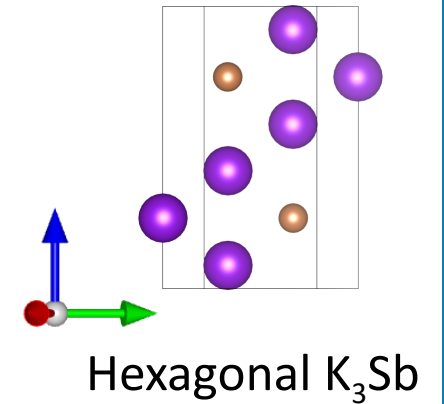
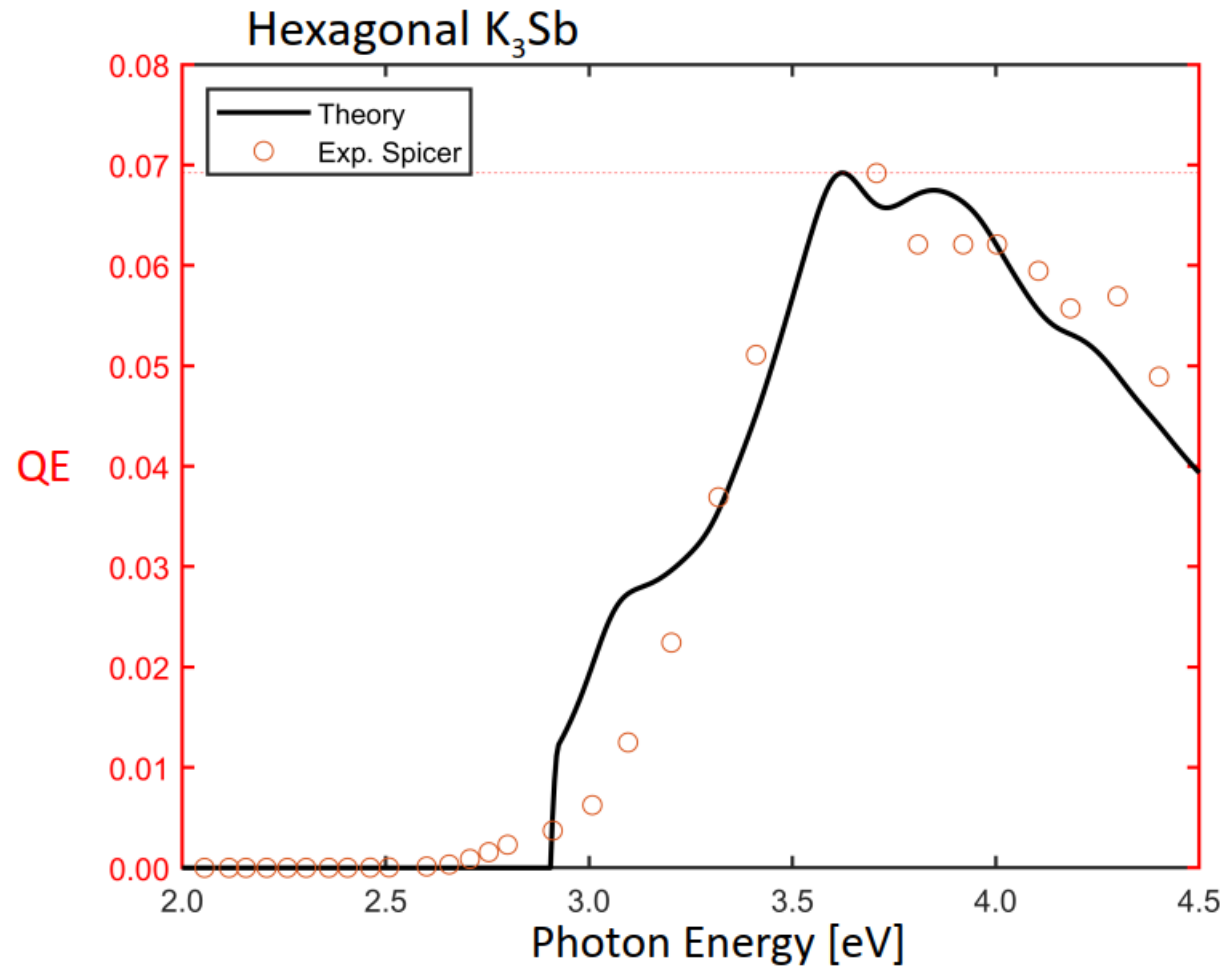
Emission by Photon Energy



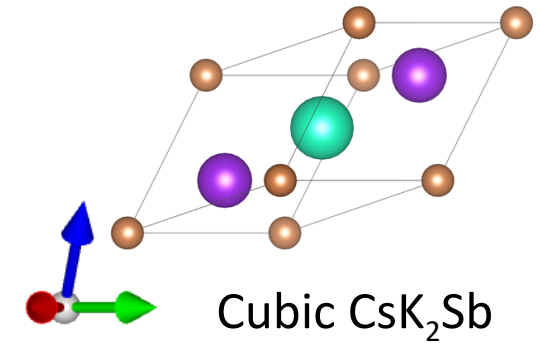
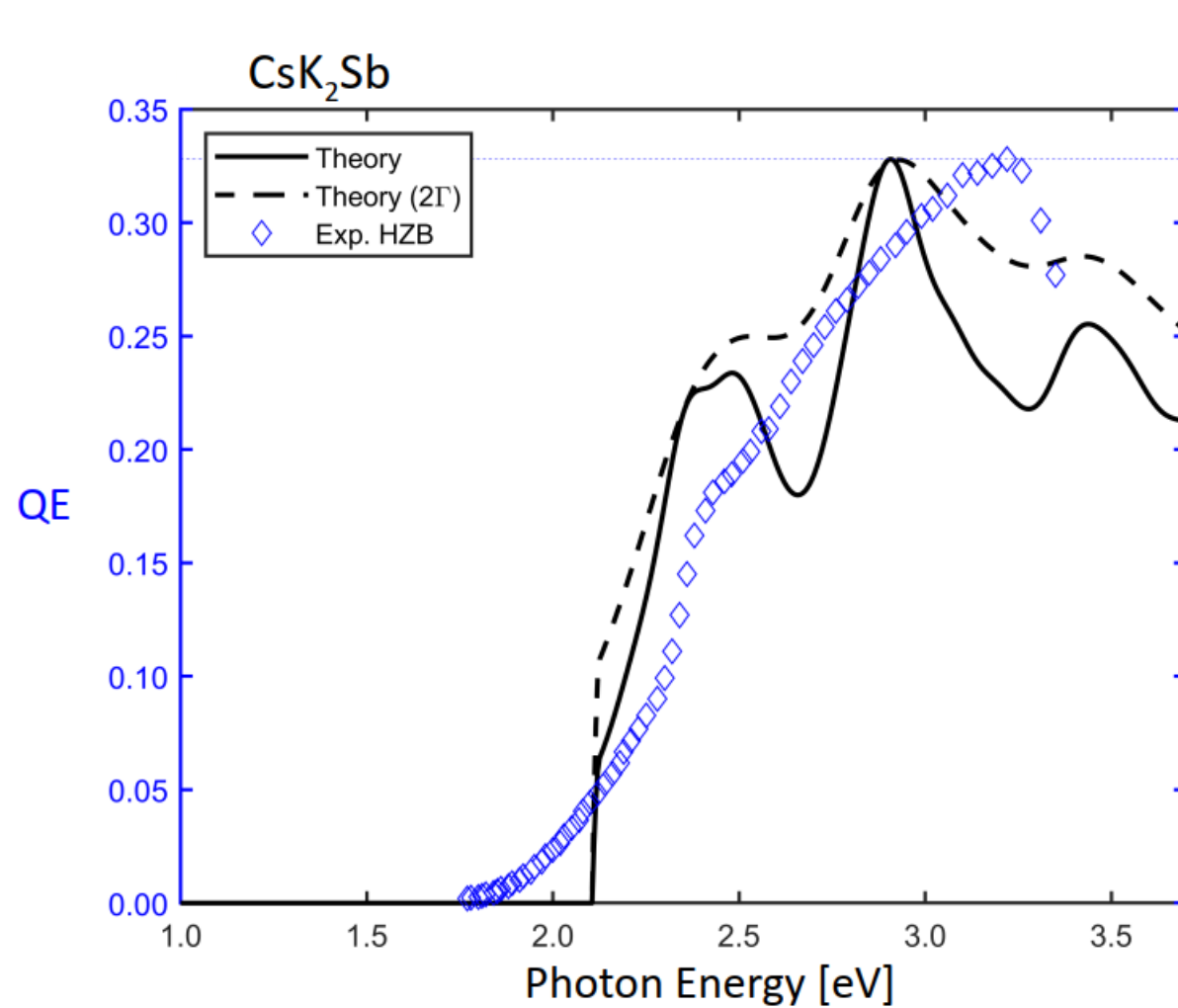
Final Result



Experimental Comparison



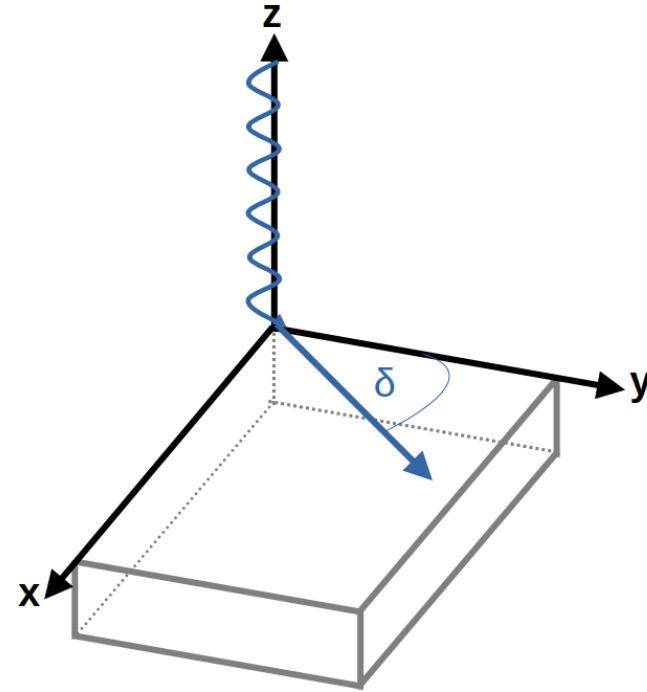
Ternary Example: CsK₂Sb



Quantitative Description: Maxwell's Equations

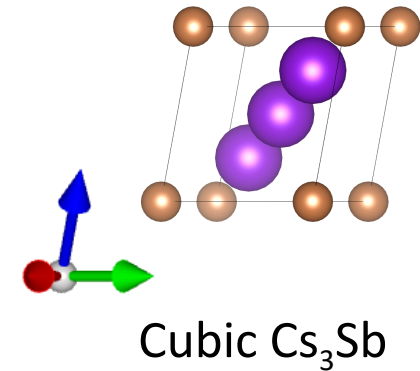
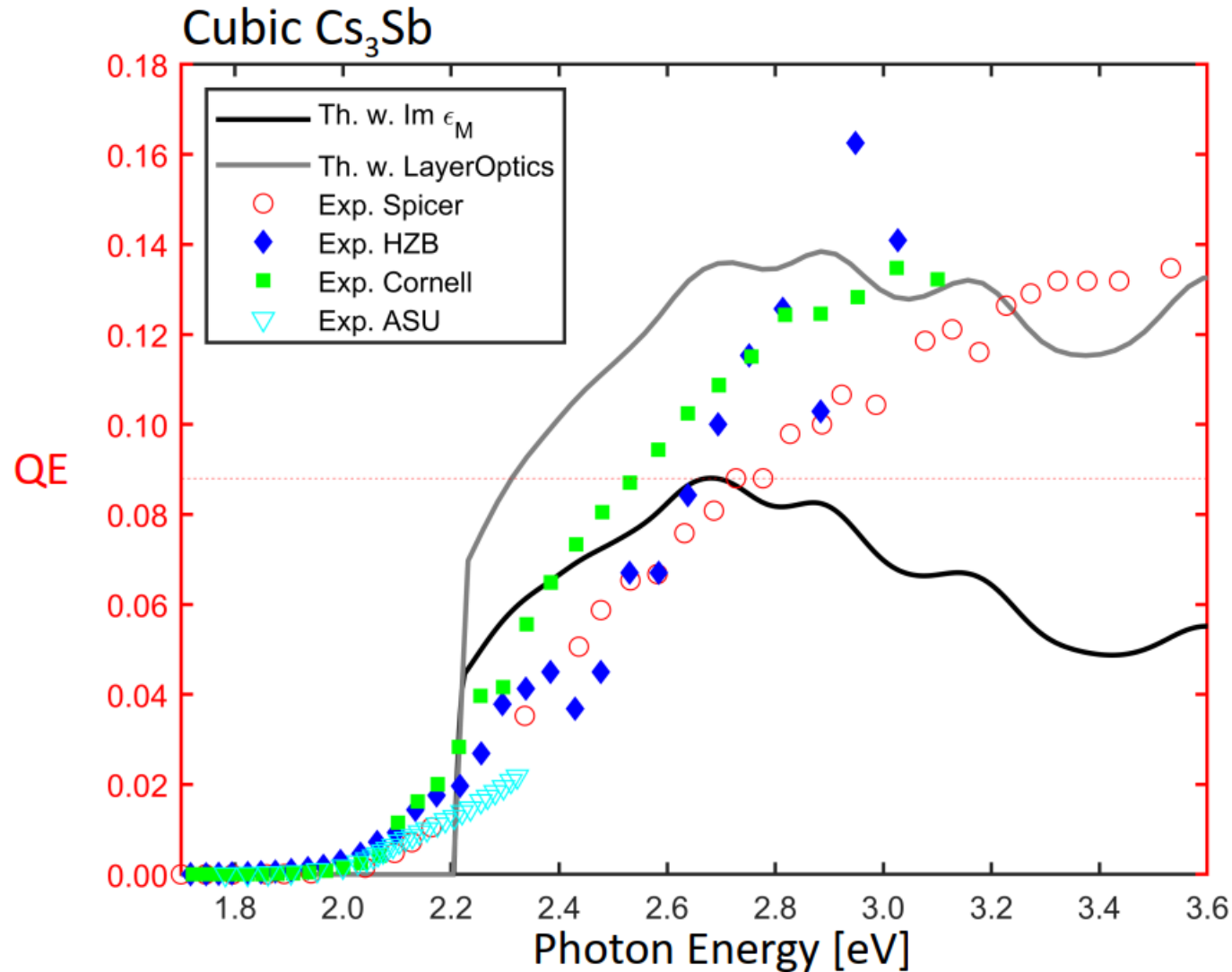
$$\vec{k} \times (\vec{k} \times \vec{E}) + \frac{\omega^2}{c^2} \epsilon(\omega) \vec{E} = 0$$

$$\vec{H} = \frac{1}{\mu_0 c} \vec{k} \times \vec{E}.$$



- ➡ Fresnel coefficients for reflectance and transmittance
- ➡ Calculated with an updated version of the code LayerOptics (C. Vorwerk et al., Comput. Phys. Commun. 201, 119 (2016))
- ➡ $\tilde{P}_{\text{Excitation}}(\omega) = 1 - (R(\omega) + T(\omega))$

Quantitative Example: Cs₃Sb



Summary

- We adopted Spicer's three-step model to create a description of the photoemission process based on the results from *ab initio* many-body theory.
- The developed model allows qualitative and quantitative predictions and has been validated against measurements for representative alkali-antimonides.
- Outlook: Quantitative improvements by incorporating inelastic scattering and explicitly modeling surface effects.
- Paper preprint:

R. Schier et al., arXiv preprint arXiv:2602.12997 (2026)