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Ab initio many-body photoemission theory of transverse momentum distributions of photoelectrons from single-crystal materials: PbTe(111) as a case study*

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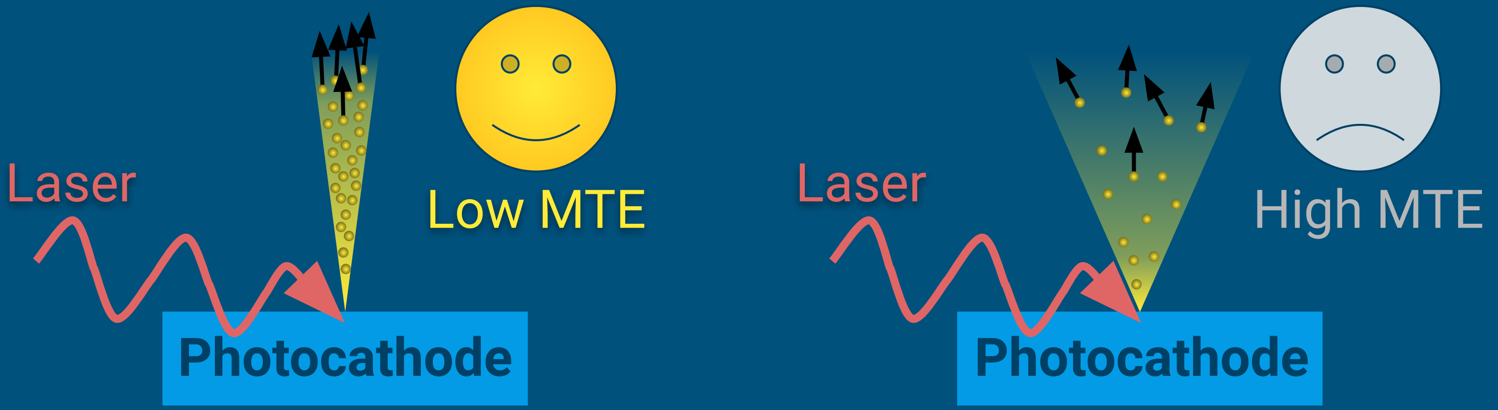
*J. K. Nangoi *et al.*, Phys. Rev. B **104**, 115132 (2021).



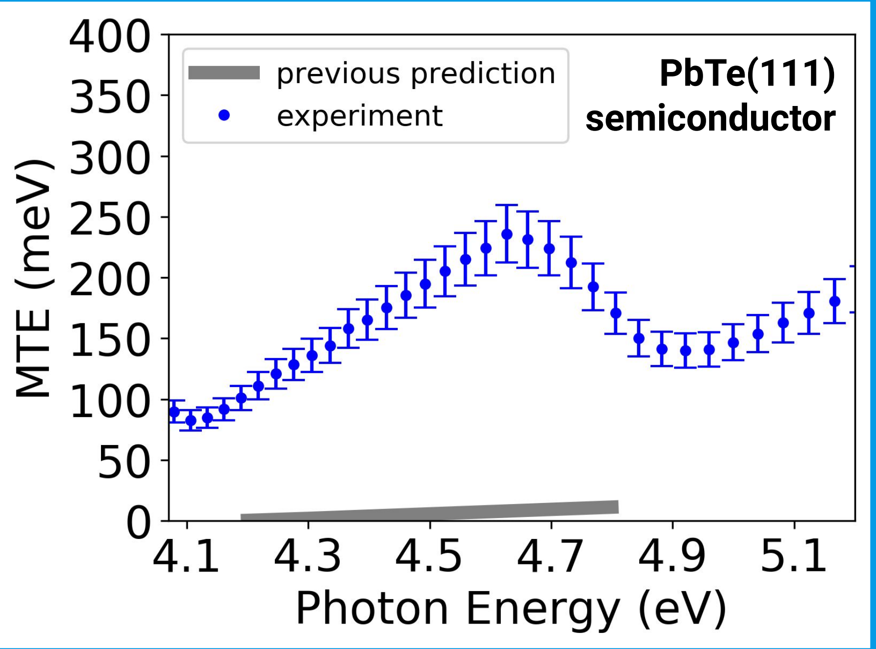
Why need *ab initio* theory?

- To understand fundamental processes relevant to MTE, e.g.
  - scattering between electrons and phonons (crystal vibrations)
  - bulk & surface effects: emission of bulk & surface electrons

Outline

- Motivation for *ab initio* studies of photoemission from PbTe(111)
- *Ab initio* bulk photoemission theory including phonon effects
- Results & interpretations, comparison with our experiments
- Further improvements to theory by including surface effects
- Summary

Motivation for PbTe(111)



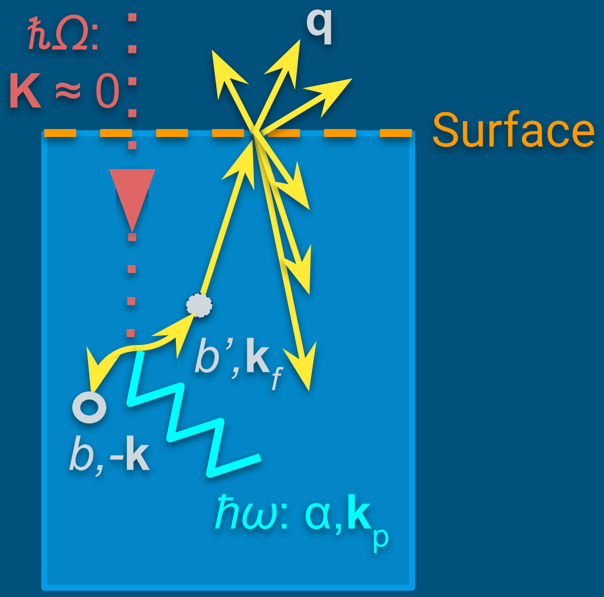
- Previous predictions* yield MTE ≤ 15 meV
  - Emission directly to vacuum
- **Our experiments 10–20× larger** and **shows photoemission below threshold**
  - Light penetration depth ~200 Å
    ⇒ bulk emission important?
  - Phonons can affect e⁻ momenta
    ⇒ phonon effects on MTE important?
- Need new *ab initio* photoemission theory including bulk emission & phonon effects

*W. A. Schroeder, T. Li, and B. Rickman, P3 2016.

Ab initio Bulk Photoemission Theory*

More bulk e^- than surface e^-

1. Photon excites bulk e^--h^+ pair at rate ν
   - Direct: photon → e^--h^+
   - Phonon-mediated: photon + phonon → e^--h^+
2. e^- in a *coherent* outgoing scattering state
   - Conservation of energy and transverse momentum gives rise to transmission probability $t(\mathbf{q})$



- Calculate photoexcitation transition rates ν and transmission probabilities t
- MTE = $\langle \hbar^2/(2m)\, q_{||}^2 \rangle$ weighted by $\nu \cdot t(\mathbf{q})$

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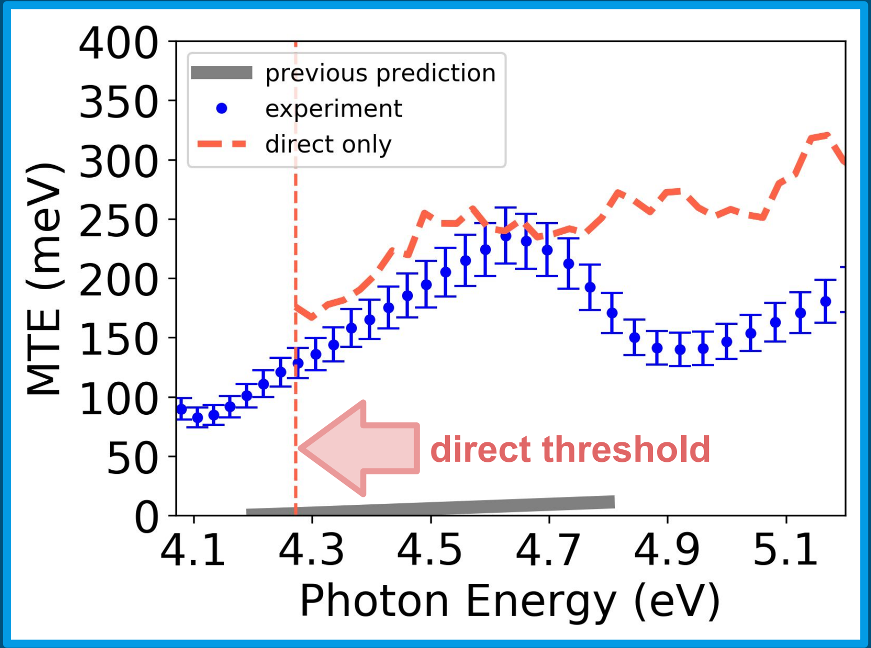
Results: Calculated MTE from PbTe(111)

- Direct only reproduces magnitude, but has higher threshold



Bulk emission important!

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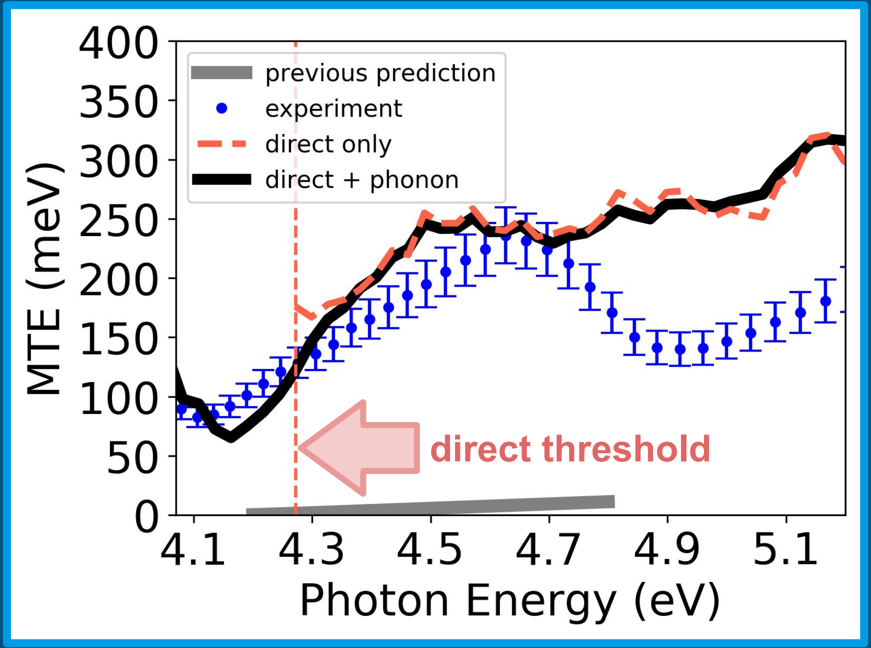
Results: Calculated MTE from PbTe(111)

- Direct + phonon reproduces photoemission below threshold



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Phonon effects important below threshold!

Results: Calculated MTE from PbTe(111)

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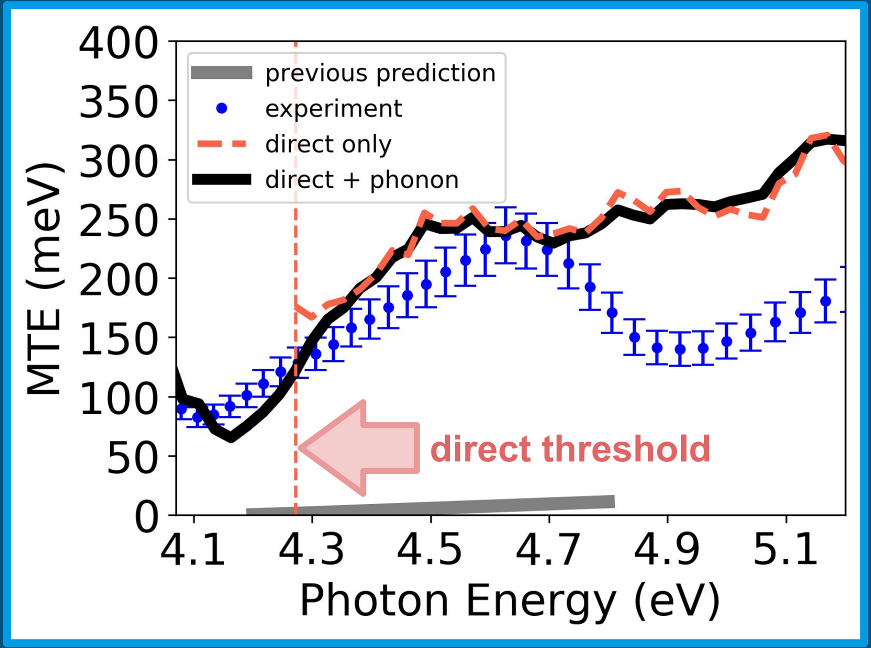

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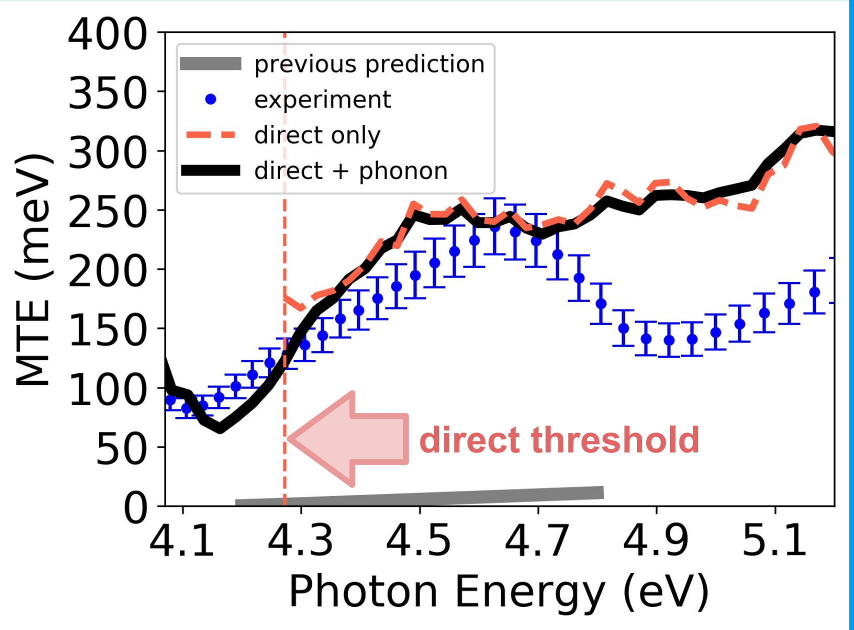
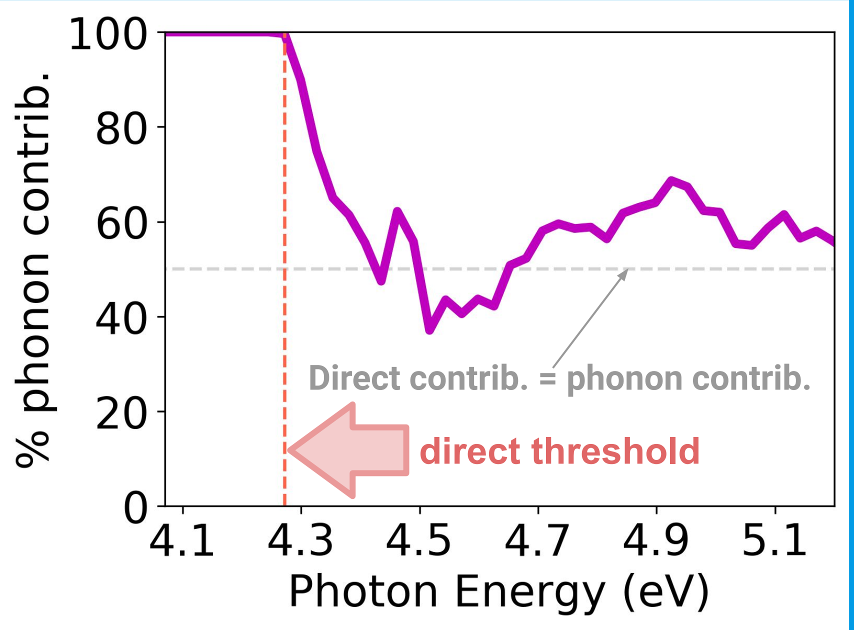
With two big questions answered, smaller questions to explore:

1. Phonon effects above direct threshold significant?
2. Reason for observed MTE dip centered at 4.9 eV?

First exploration: Significance of phonon effects

- Phonon effects above direct threshold ***significant!***





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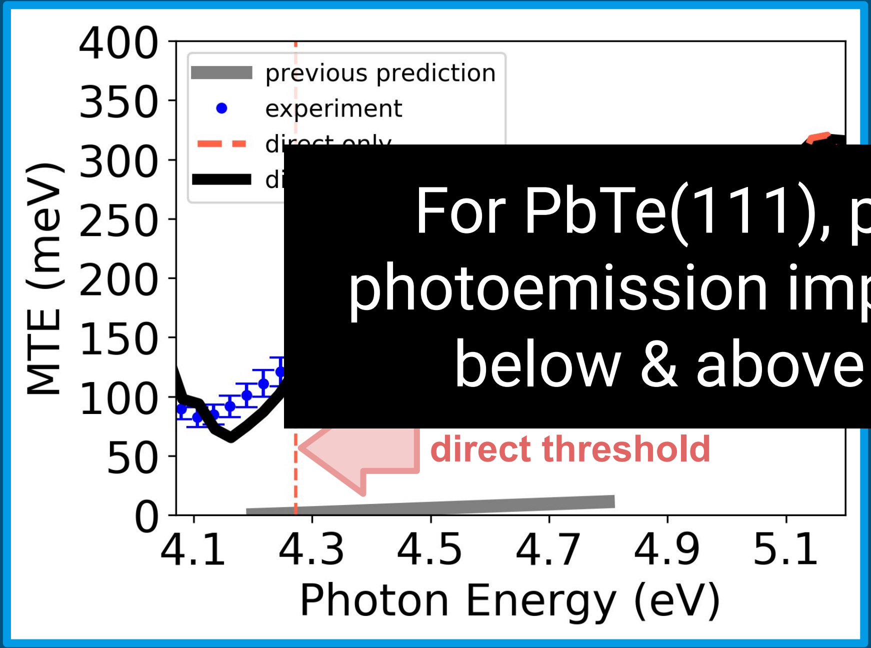

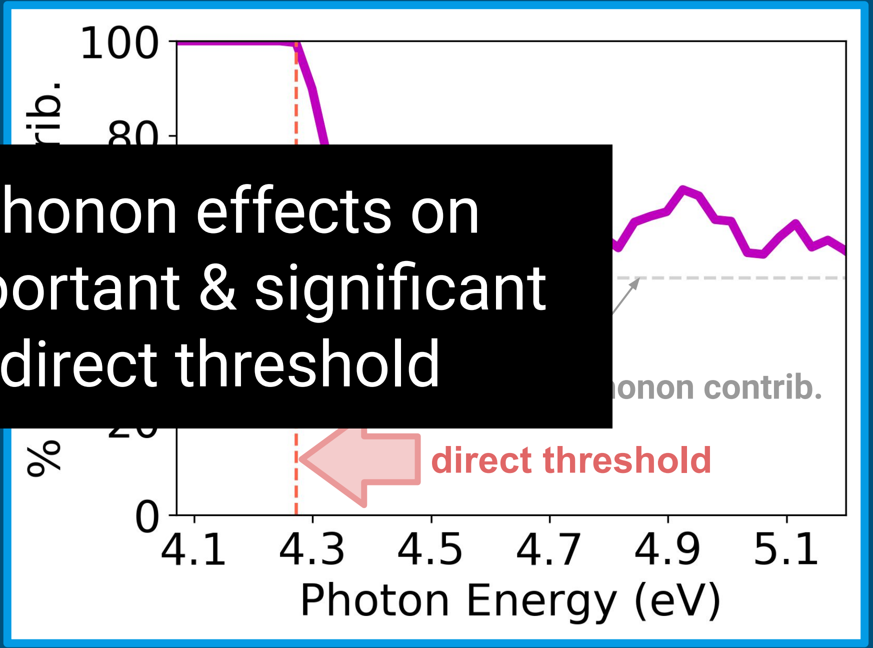

For PbTe(111), phonon effects on photoemission important & significant below & above direct threshold

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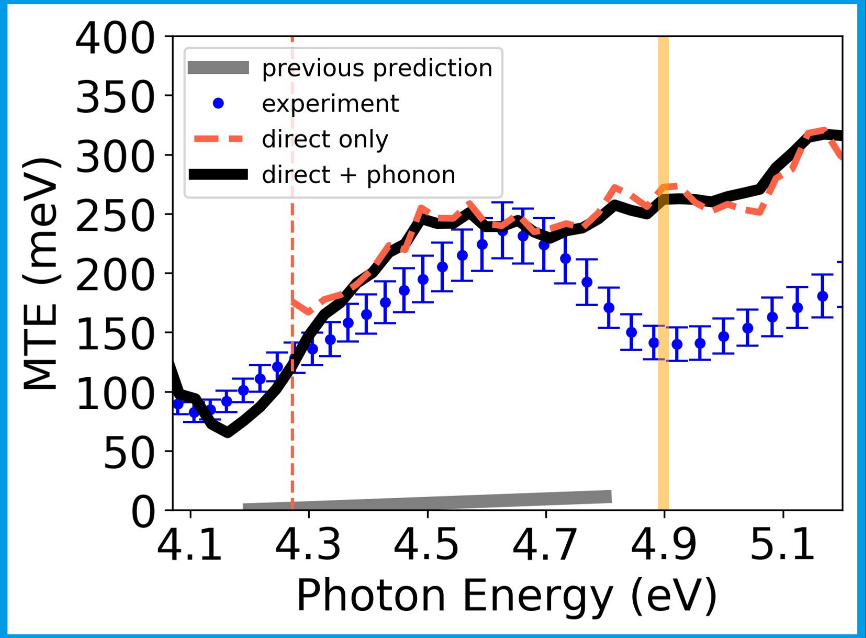
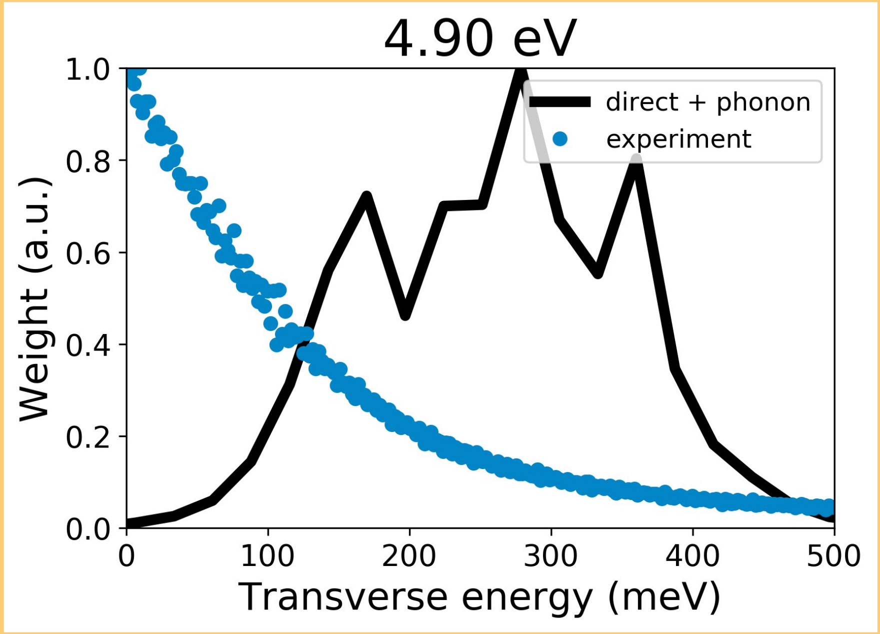
Second exploration: MTE dip at 4.9 eV

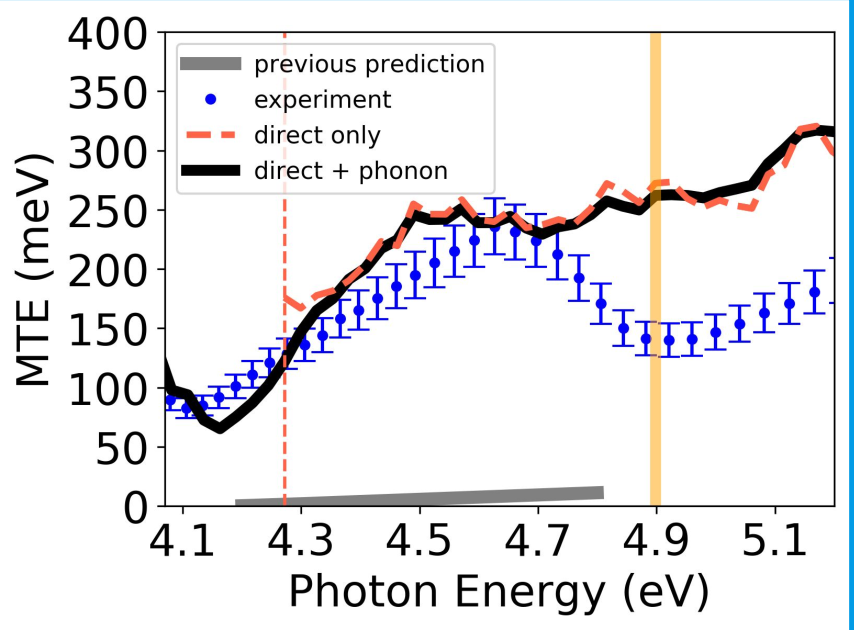
Calculations include emission of only bulk electrons

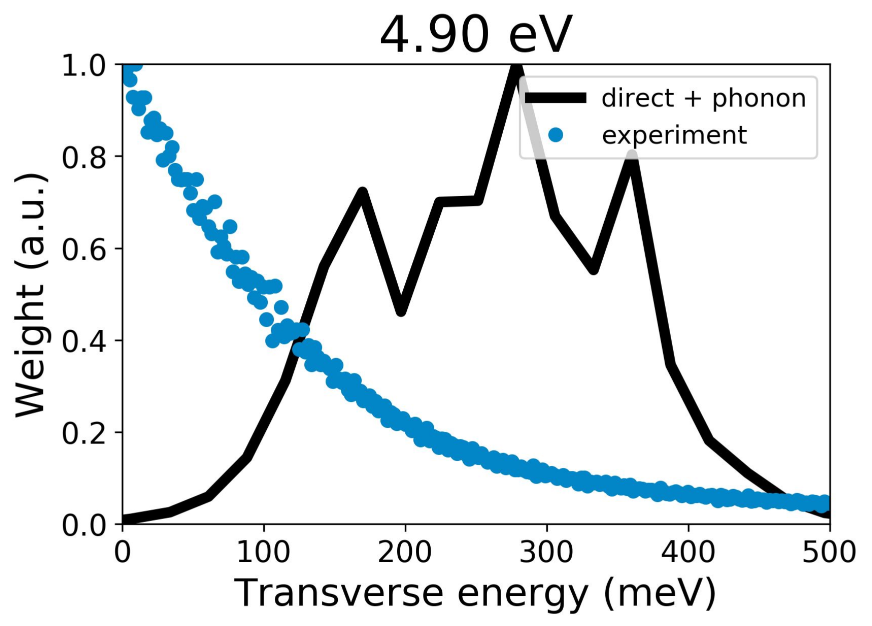




Second exploration: MTE dip at 4.9 eV

⇒ Contributions at 0 TE likely due to emission of *surface* electrons





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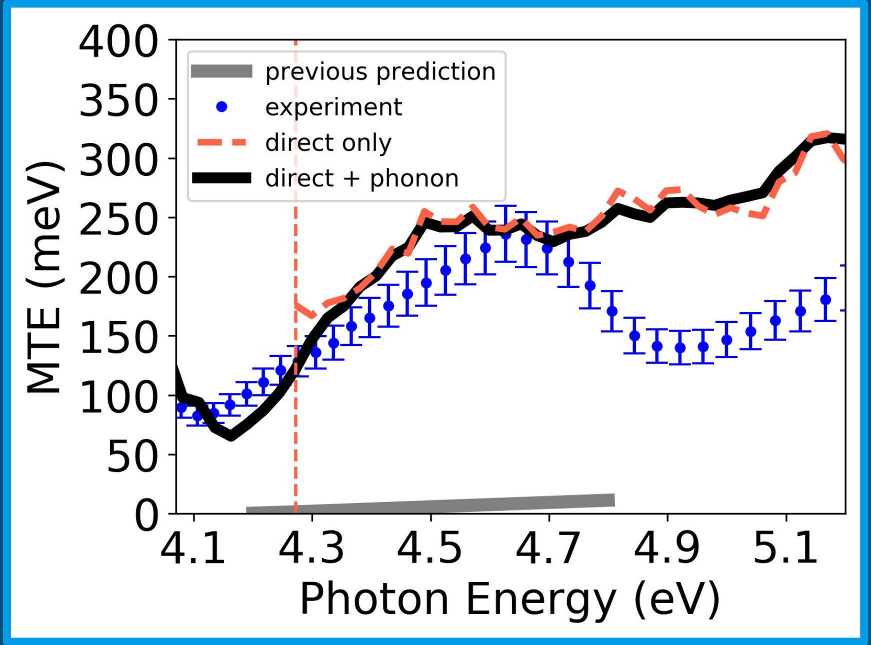

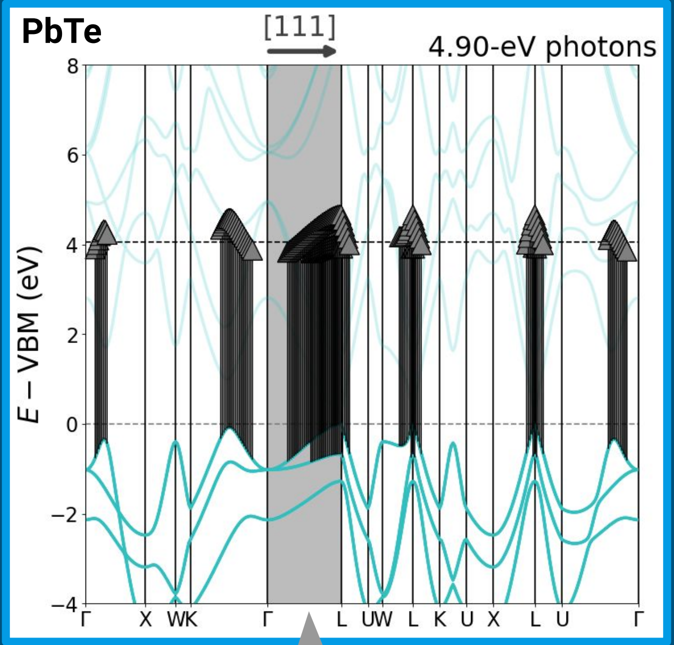

Vacuum level

Valence band maximum (VBM)

Zero transverse energy

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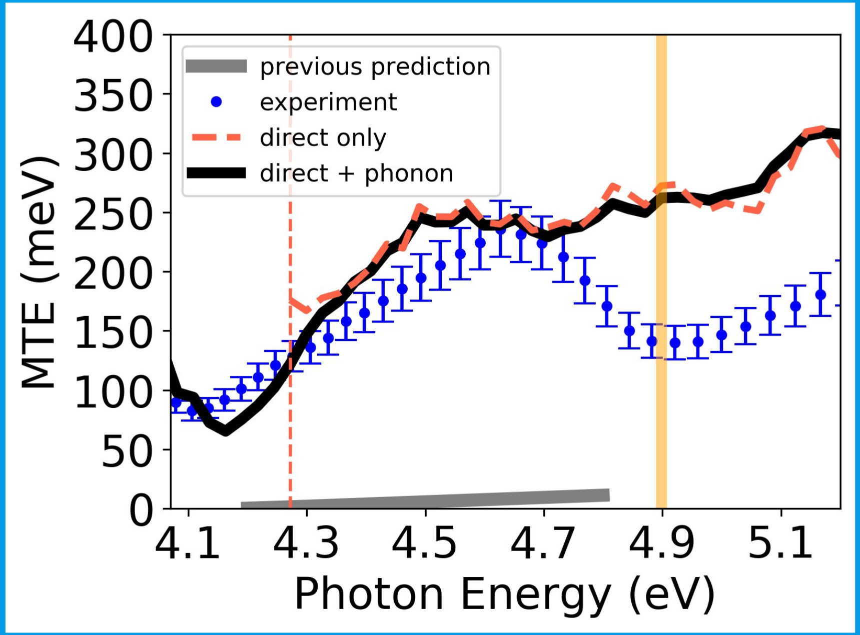

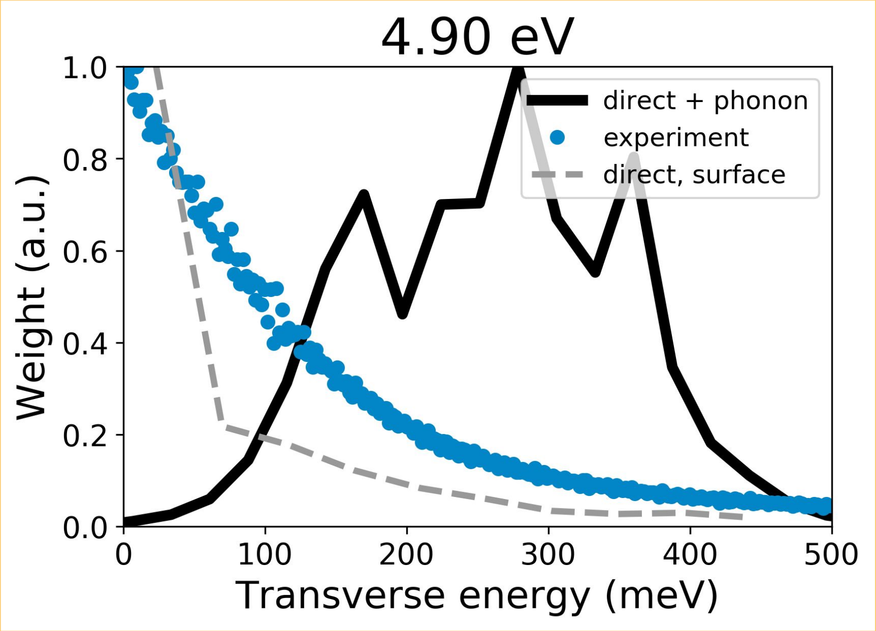

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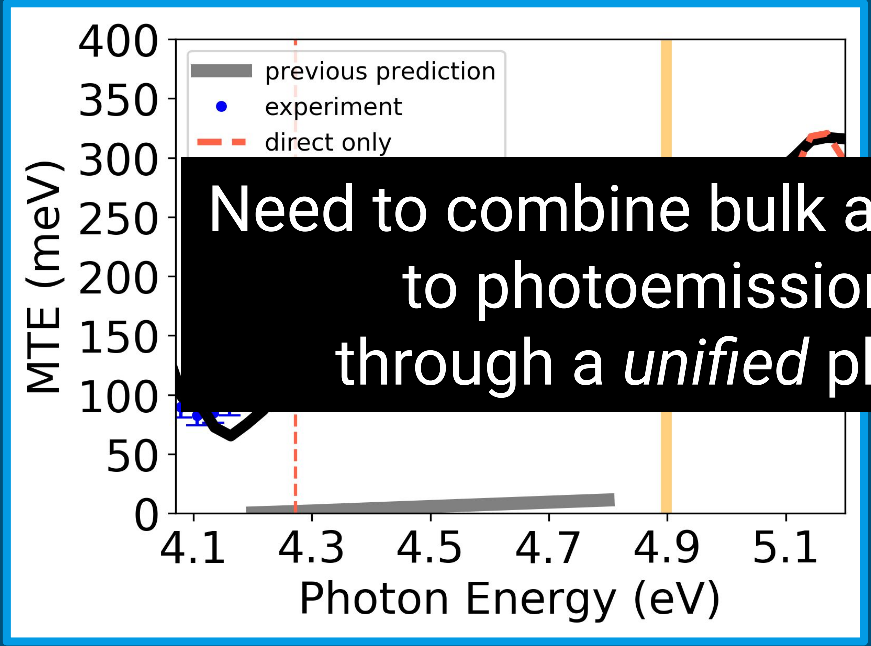

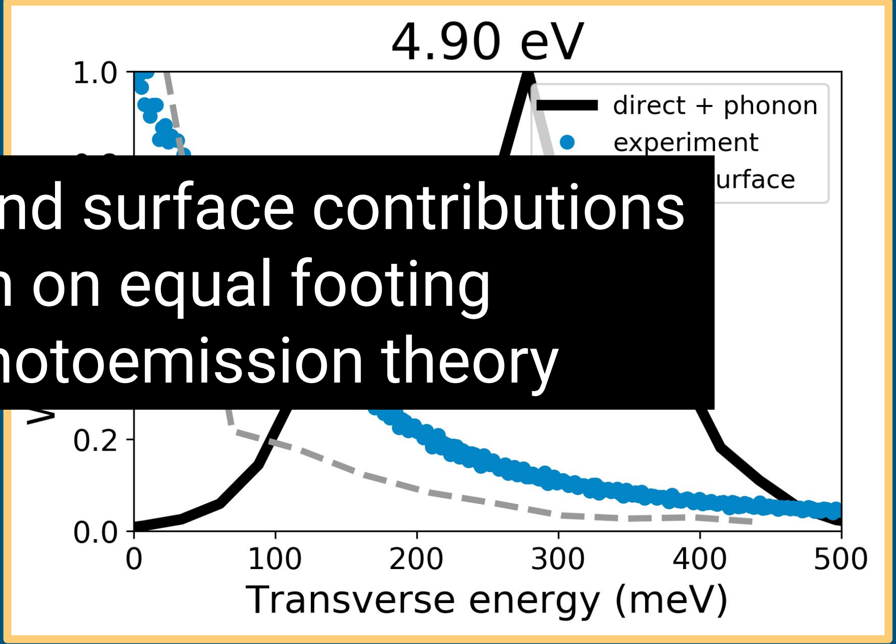

Need to combine bulk and surface contributions to photoemission on equal footing through a *unified* photoemission theory

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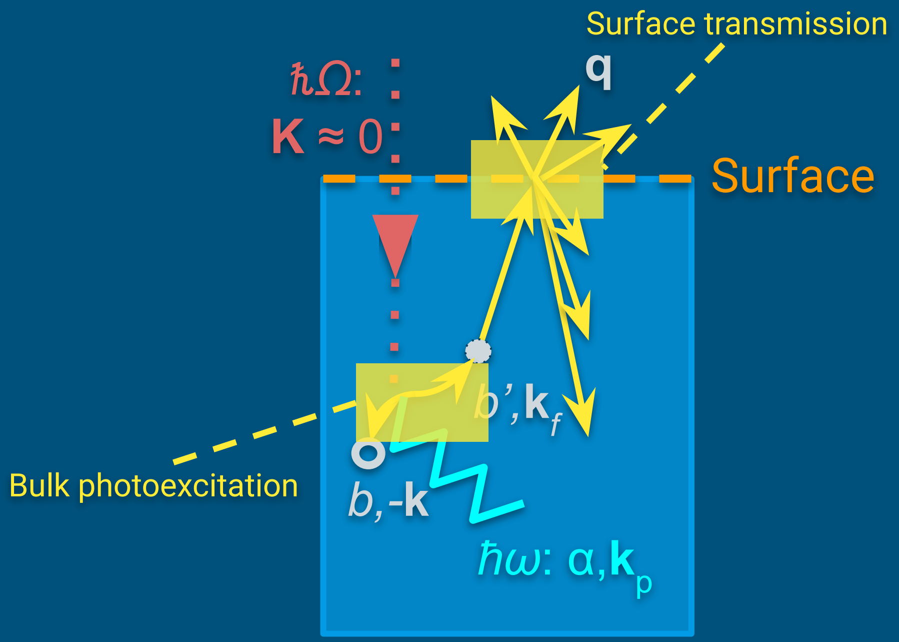
Unified Photoemission Theory: One-step Model*

Instead of treating photoexcitation and surface transmission separately …

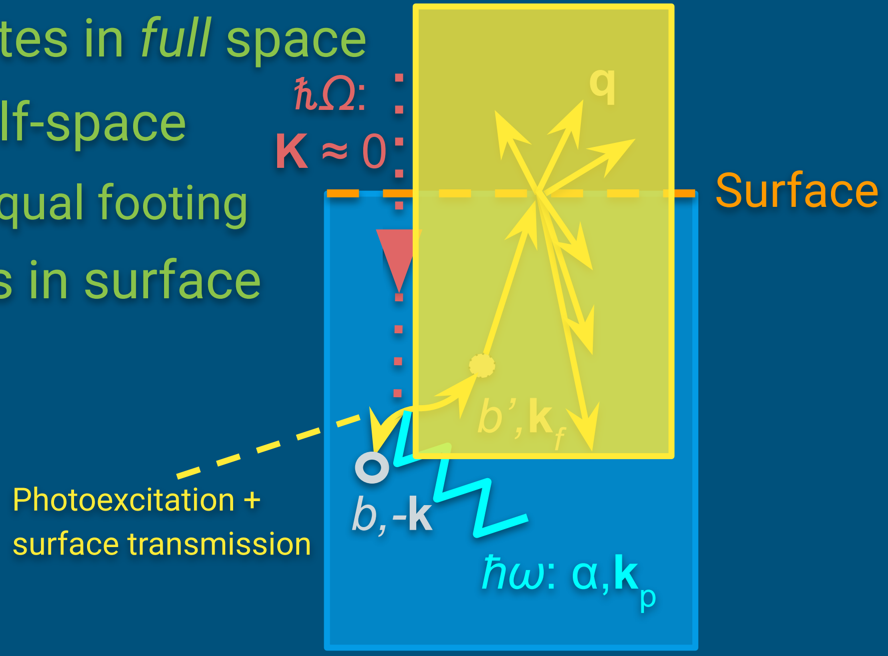


* e.g. P. J. Feibelman and D. E. Eastman, Phys. Rev. B **10**, 4932 (1974).

Unified Photoemission Theory: One-step Model*

... treat them both ***simultaneously*** in a single step

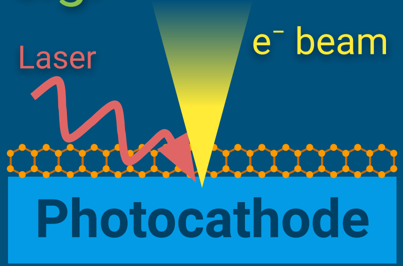
- Requires expressing electronic states in *full* space = material half-space + vacuum half-space
  - Bulk, surface, vacuum states on equal footing
- *Challenge*: non-periodic boundaries in surface normal direction



* e.g. P. J. Feibelman and D. E. Eastman, Phys. Rev. B **10**, 4932 (1974).

New *ab initio* one-step photoemission theory

- Developed new technique combining electron Green's function approach with plane-wave DFT to handle non-periodic boundaries
- Currently implementing direct photoemission; will implement phonon-mediated photoemission
  - Then, apply to PbTe(111) and others e.g. Cs-Sb, Cs-GaAs, etc.
- Ultimately, plan to study photoemission from e.g. ***photocathodes coated with 2D materials*** (talks in Session A yesterday)



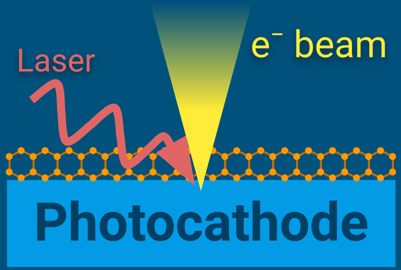
Summary

- *Ab initio* many-body photoemission theory for predicting MTE
- Case study on single-crystal PbTe(111):
  - Calculated MTEs same/similar magnitude as observed MTEs
  - Phonon effects important & significant
  - Surface emission seems significant at higher photon energies

    ⇒ *Need to include bulk and surface contributions on equal footing*

Developing new *ab initio* one-step photoemission theory to include bulk and surface emissions in a unified framework

- Plan to study photocathodes coated with 2D materials



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