Ab initio theory of coherent electron-photon-phonon scattering during photoemission

transverse momentum distribution of electrons from PbTe(111) photocathodes as a case study

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High-brightness photocathodes require low MTE (< ~10 meV)
MTE ≡ mean transverse energy ∝ ⟨|transverse momentum|2⟩

Next-generation applications of low-MTE photocathodes:
- XFEL with shorter length (~1 km → ~100 m)
- UED with larger maximum size of system under study
- More accurate studies of:
  - Ultrafast photo-induced metal-insulator transition of VO2
  - Ultrafast photo-conversion dynamics in rhodopsin

Expect phonon scattering to affect MTE, but to date no fully ab initio photonemission theory with full inclusion of phonon effects

So, develop ab initio photonemission theory including coherent three-body electron-photon-phonon scattering to calculate MTE

Ab initio calculations of MTE

Direct photoexcitation
\[ \nu_{k',b'}^{\text{direct}} = \frac{2\pi}{\hbar} \frac{e^2}{m^2} A_0(\Omega) (f_{k,b} - f_{k',b'}) \times \delta (E_{k',b'} - E_{k,b} - \hbar \Omega) \times |\hat{e}(\Omega) \cdot \mathbf{p}_{k',b' + b}|^2 \]

Phonon-mediated photoexcitation
\[ \nu_{k',b' + b}^{\text{phonon}} = \frac{2\pi}{\hbar} \frac{e^2}{m^2} A_0(\Omega) (f_{k,b} - f_{k',b'}) \times \sum_{\alpha=\pm} \left( \frac{n_{k' - k,\alpha}}{g_{k' - k,\alpha}} + \frac{1}{2} \right) \times \delta (E_{k',b'} - E_{k,b} - \hbar \Omega + \hbar \omega_{k' - k,\alpha}) \times |\hat{e}(\Omega) \cdot \mathbf{p}_{k',b' + b}|^2 \]

Transmission into vacuum
\[ t(k_f, b', G_1) = \Theta (\nu_{\text{group}, \alpha} \cdot \hat{n}) \Theta (\mathbf{T}_L^{k_f, b', G_1}) \times |D_{k_f, b'} G_1|^2 (2/m_c) \mathbf{T}_L^{k_f, b', G_1}^{1/2} \]

Monte Carlo integration using Wannier interpolation

- Calculate matrix elements and linewidths in Wannier basis
- Interpolate to arbitrary \( k_f \) and sum over all \( k_f \)'s in BZ1 using Monte Carlo integration

Results: Photoemission from PbTe(111)

Conclusions & Future Work

- Bulk phonon-mediated photoemission important on PbTe(111)
  - Indirect photoemission below direct threshold detected in our experiments
  - Significant phonon-mediated photocurrents above direct threshold even at 30 K
- Our calculations reproduce our experiments far better than previous prediction
- Future work: Solve remaining discrepancies by using ab initio one-step photoemission