First-principles many-body study of the electronic and optical properties of CsK₂Sb, a semiconducting material for ultra-bright electron sources

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• Electronic-structure characterization of CsK₂Sb as photocathode material for particle accelerators with state-of-the-art ab initio methods
• Compute the relevant quantities, such as band structure, band character, and optical absorption that are needed to model quantum efficiency (QE) and intrinsic emittance (IE)
• Theoretical core spectroscopy for further characterization
• First step towards advanced modeling of photoemission properties of photocathode materials

CsK₂Sb as photocathode material

Excellent photocathode characteristics [1]:
• Band gap and electron affinity of ~1 eV [2,3]
• Photoemission close to threshold in the near-infrared and visible band
• High QE in the visible region
• Robustness
• Low dark current
• Short response time

Goals

Methodology

First-principles approach based on density-functional theory (DFT) and many-body perturbation theory (MBPT)

DFT

• Mean-field approach
• Kohn-Sham equations [4]

G₀W₀

• Electron-electron correlation
• Hedin’s equations [5]

BSE

• Electron-hole correlation
• Two-particle Bethe-Salpeter equation [6]

Electronic structure

• Band gap of 1.62 eV from G₀W₀@DFT.
• DFT alone underestimates the gap by about 0.7 eV.
• Valence region characterized by flat p-like bands with predominant Sb contributions
• Conduction-band bottom with parabolic s-like bands, exhibiting strong hybridization between Sb and Cs atoms.
• Contributions from K atoms appear mainly at higher energies in the conduction region with d-like states

Optical absorption

• Absorption in the visible region; excitonic effects red-shift the peaks by ~0.2 eV and redistribute the oscillator strength towards lower energies.
• Bright optical excitations up to 2.5 eV (A, B, and C) stemming from transitions between the valence-band top and the conduction-band bottom at Γ; peak at 3 eV from transitions at X.
• Plasmonic excitations given by maxima in the loss functions at about 5.5 eV and 11 eV

Core spectroscopy

• X-ray absorption spectrum dominated by a strong peak at the absorption onset.
• Excitonic nature of the first peak: absent in the independent-particle (IPA) non-interacting picture.
• The first excitation (vertical line) is 4-fold degenerate with contributions from 2 different types of transitions: At lowest energy transition to the te Γ-point (Cs-s character) while a few meV above transition to the Cs d-state at the X-point.

Conclusions and Outlook

• Ab initio many-body characterization of the electronic and optical properties of CsK₂Sb
• Band gap and optical absorption onset in the visible region, at about 1.5 eV
• Weakly dispersive valence band with predominant Sb p-like character
• Coexisting s- and d-like contributions in the conduction region: highly dispersive bands
• Comprehensive first-principles study of multi-alkali antimonides as novel materials for ultra-bright electron sources
• Improved modelling of photoemission based on ab initio results

References


C. Cocchi et al., arXiv1809.00135 (2018)