MULTISCALE MODELING OF INTERFACES PHOTOVOLTAIC DEVICES

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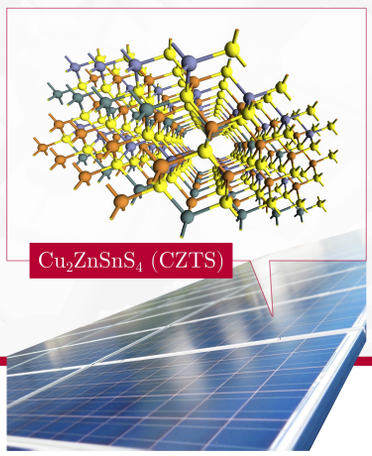
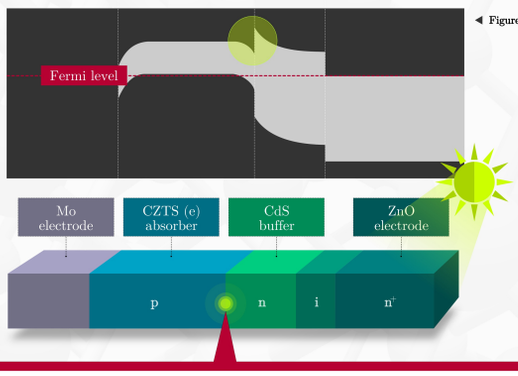
Abstract: We present an alternative explanation for the open circuit voltages (OCV) deficit in Cu_2ZnSnS_4 (CZTS). Using a method[1] based on tools available in the Atomistix Tool Kit (ATK) software package, we have studied the electronic structure at the interface between CZTS and CdS in detail. Here, we have identified a shallow state localized at the interface. When included in device level simulations, such a state leads to significant deterioration of the OCV, and when this is taken into account, we can quantitatively reproduce measurements on state-of-the-art CZTS solar cells. This shows how parameters needed for device characterization can be extracted from atomistic device simulations and how the interplay between atomistic and device level simulations can be a powerful tool in characterizing the thin-film solarcell devices of the future.

Solar Cell: CZTS|CdS Interface





◀ **Figure 1|** Schematic of the full cell showing the approximate band structure.

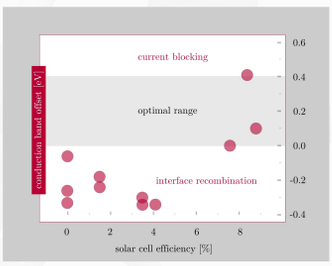


Figure 2| Solar cell efficiency vs. measured CBO value for CZTS/CdS interface.

The full solar cell consists of several layers of materials, with loss mechanisms related to each of them, as well as the interfaces between them. In this work, we focus on the interface between CZTS(e) and CdS, which has been shown to be important for this class of materials.

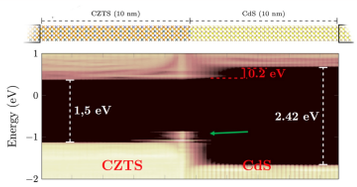


Figure 3| Band structure at the CZTS/CdS interface.

The efficiency is optimized when the CBO value is between 0 and 0.4 eV, which is consistently both calculated and measured to be the case for CZTS/CdS.

We show that, under flat-band conditions, the CBO is 0.2 eV, which is right in the middle of the optimal range. However, we also show that there are interface states in the gap, effectively lowering the band gap.

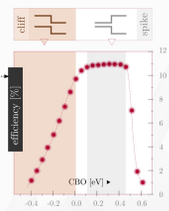


Figure 4| Calculated efficiency as a function of CBO.

Modeling the Device with DFT-NEGF

Figure 5| The device model being used for atomistic simulations with the DFT-NEGF method. Note the scale.

We use the DFT-NEGF method implemented in the Atomistix ToolKit (ATK) software package to calculate the band structure of an interface between two semi-infinite bulk materials. This allows us to treat the interface much more realistically then previous works, which uses true periodic bulks, meaning that the interface was infinitely repeated. In our case, there is only the one interface in the system, which is coupled to true bulk regions of CZTS and CdS.

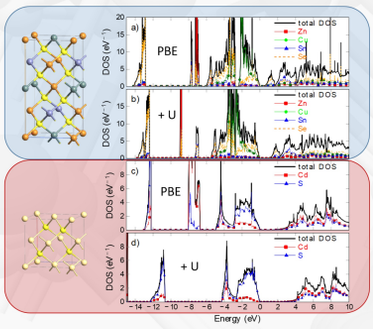
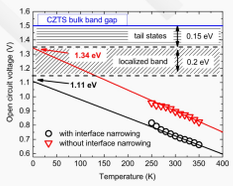


Figure 6| DOS for the two materials, with and without the +U correction. The correction clearly opens up the band gap, as expected.

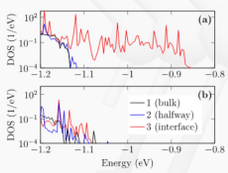
Calculating Circuit Voltage

In order to find the macroscopic effect of these interface states, we performed device-level simulations using the SCAPS software. As expected, this showed that the open-circuit voltage was significantly lowered by the interface states, and the value at room temperature, 720 mV, is very close to the experimental record of 710 mV. This indicates that these interface states are the cause of the lower than expected efficiency for even the best CZTS/CdS solar cells.

Figure 7| Calculated open circuit voltage as a function of temperature, with and without inclusion of the interface states. ▶



Results



After further analysis, it was determined that ZnS would be a good candidate for a buffer material which could remove the interface states, and thereby increase the maximum efficiency of the solar cell. The figure shows that the interface states are indeed gone when CdS is exchanged with ZnS.

Figure 8| Close-up of the DOS near the VBM for the CZTS/CdS (top) and CZTS/ZnS (bottom) interfaces.

Conclusions

- We have presented a multi-scale framework for simulating PhotoVoltaics.
- Device parameters, such as band offsets, are calculated from first principles using DFT-NEGF with the +U correction and included in a drift-diffusion device simulator – in this case the SCAPS-1D software
- Applying this approach, we obtain quantitative results for a CZTS/CdS device, which shows the interface states in the gap. They can be included in the drift-diffusion model, which reveals the importance of interface states.

References

[1] G. Margaritondo, Electronic Structure of semiconductor heterojunctions, (1988).

VIRTUAL NANOLAB — ATOMISTIX TOOLKIT