

Pressure on solar cells

Calculation for high-pressure-behaviour of solar cell materials

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In Short

- Kesterites ($\text{Cu}_2\text{ZnSnS}_4$) are a promising material class for a cost-effective and distributed electrical power production with thin film solar cells from solar radiation.
- The structural and electronic properties of $\text{Cu}_2\text{ZnSnS}_4$ bulk systems have been theoretically studied extensively, showing good agreement with experimental data regarding structural and electronic properties.
- There are only few theoretical studies on the high pressure phases of $\text{Cu}_2\text{ZnSnS}_4$.
- We aim to verify the published high-pressure results on the stannite (ST), primitive mixed Cu-Au (PMCA) and rock salt (NaCl) structure. Furthermore we want to consider additional possible structures.
- For each phase we will carry out a volume scan to identify possible phase transitions and calculate the density of states (DOS) to get insight into the electronic properties.

The impending exhaustion of fossil fuel has prompted the exploration and exploitation of alternative energy resources, with the solar energy harvesting through photovoltaic devices spearheading these efforts. In an attempt to overcome the restraints of Si-based materials, the direct optical band gap of chalcogenide-based solar cells offers the benefit of higher absorption in comparison to silicon. Given that the employed chalcogenides are composed of multiple elements, one can additionally optimize the photovoltaic properties of the respective material by appropriate metal or chalcogenide substitution. Among the various chalcogenides investigated for this purpose, the quaternary semiconductor $\text{Cu}_2\text{ZnSnS}_4$ has attracted considerable attention in recent years. [1,2] The suitability of this material for solar cell applications stems from its almost optimal band gap ($E_g \approx 1.5$ eV), its high absorption coefficient in the visible range (maximum around 1000 nm), and its earth-abundant, low-cost, and non-toxic constituents. [3]

At ambient conditions, $\text{Cu}_2\text{ZnSnS}_4$ crystallizes in the tetragonal kesterite (KS) structure (SG $I\bar{4}$, $Z = 2$, Fig. 1). [4] This phase, which is structurally related

to the sphalerite- and chalcopyrite-type structures, is composed of alternating cationic (Cu/Sn or Cu/Zn) layers along the c -axis interrupted by sulfur anions, with all of the cations tetrahedrally coordinated with respect to anions. Even though the KS phase represents an ordered cationic arrangement, i.e. each cation occupies a unique Wyckoff site, cationic disorder is quite common in this material. [4,5] Such disorder can affect the photovoltaic properties of this system. For example, a mixed occupancy between Cu and Zn in the $z = 1/4$ and $z = 3/4$ cationic layers results in a disordered kesterite (DKS) configuration (SG $I\bar{4}2m$, $Z = 2$, Fig. 1) and a reduction of E_g . [6]

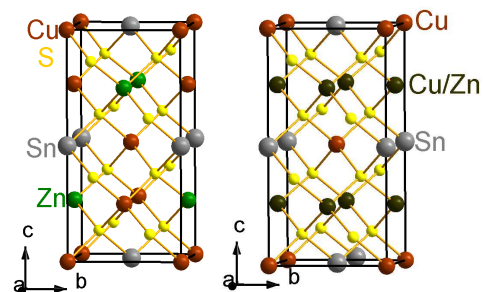


Figure 1: Unit cell of the kesterite-type $\text{Cu}_2\text{ZnSnS}_4$ (KS, SG $I\bar{4}$, $Z = 2$, left) and the disordered-KS (DKS, SG $I\bar{4}2m$, $Z = 2$, right) phases. The red, green, gray, and yellow spheres represent Cu, Zn, Sn, and S ions, respectively; the dark spheres in the DKS phase depict the mixed Cu/Zn occupancy.

A vast amount of the theoretical work was performed on the kesterite structure. To investigate effects under pressure also other phases e.g. stannite (ST), primitive mixed Cu-Au (PMCA), or rock salt-type structure can occur, where only very little, mainly theoretical, work is available. [7]

Within this project we plan to calculate the phase diagram for structures mentioned above, by carrying out a volume scan. We will additionally consider other possible structures. To gain insight into the electronic properties we will calculate the density of states (DOS) for each phase.

In our computational studies, the electronic and structural properties of the systems will be obtained by periodic density functional theory calculations. In a first step we use the GGA functional PBE for the structure optimisations in the volume scan to reduce computational costs. The PBE functional yields sufficiently accurate results for the structural parameters in comparison to experimental data. Since the PBE functional fails to accurately describe the electronic structure, we use the hybrid functional HSE06 for

the electronic properties. The calculations will be performed with a plane wave basis set using the Vienna Ab Initio Simulation Package [8], which is known to benefit from the parallelisation over several nodes and hundred processors with sufficiently large unit cells. Only the availability of computational resources like the ones provided by HLRN allows us to investigate the described systems.

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More Information

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