Theoretical investigation of thin film solar cell materials

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Thin film solar cells based on CuIn1-xGaxSe2 (CIGSe) and Cu2ZnSnSe4 (CZTSe) are manufactured and deployed worldwide because of their outstanding price/performance ratio. Because of the complex nature of these thin film solar cell devices, it is rather cumbersome to systematically improve their efficiency. Therefore, the focus of our project is to theoretically investigate via density functional theory (DFT) calculations the atomic and electronic structure of the constituent materials involved in the production of thin film solar cells.

DFT study of the Cd and Cu exchange between the light absorber and CdS buffer

Perhaps one of the most important topics in the field of thin film solar cells is the interaction between the CIGSe and CZTSe light absorber material and the buffer layer, which is either a CdS layer deposited by a wet chemical process or a zinc-oxysulfide Zn(O,S) layer. To investigate the properties of the $CuInSe_2$, $CuInS_2$, $CuGaSe_2$, and $CuIn_5Se_8$ solar cell materials (see Fig. 1) in the presence of Cd and Cu impurities, we performed large-scale DFT calculations [1].

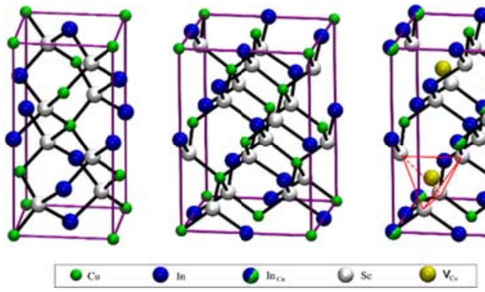

Fig. 1: Conventional tetragonal unit cell of $CuInSe_2$ (left). The tetragonal chalcopyrite-like $CuIn_5Se_8$ ordered vacancy compound (right) can be structurally understood as taking a √2×√2×1 supercell of $CuInSe_2$ (show in the center) and substituting two Cu atoms with In (denoted as In_{Cu}) while four Cu atoms are removed, leading to four pristine Cu-vacancy sites denoted as V_{Cu}.

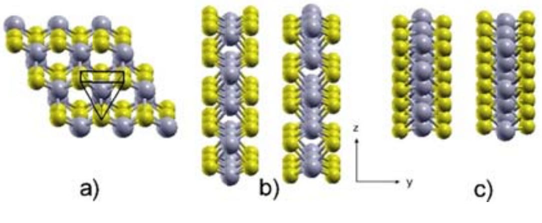

Fig. 2: Crystal structure of bulk hexagonal $MoSe_2$ (a) together with the $(10\bar{1}0)$ (b) and $(10\bar{2}0)$ surfaces (c).

Our results show agreement with recent experimental investigations, where Cd impurities are accommodated in the material, preferably in the Cu sublattice and in pristine Cu vacancy sites. The inclusion of Cd makes the $CuIn_5Se_8$ even more Cu-poor. Furthermore, by the means of ab-initio molecular dynamics, we simulated the mass transport of Cd impurities in the Cu-poor $CuIn_5Se_8$ compound [2]. We found a double-hump profile, indicating that the diffusion of Cd involves the migration of Cu as well, where Cu is exchanged between pristine defects present in the Cu-poor material. In addition to Cu, Zn, and Cd impurities, currently we are looking into the effect of Li, Na, and K dopants on the atomic and electronic structures of these materials, so that we can assess the benign nature of post-deposition treatment with alkali metals and their salts upon the improved efficiency of thin film solar cells [3]. Since grain boundaries (GBs) play a major role in the device efficiency, we also studied the effect of GBs in $CuInSe_2$ and $CuGaSe_2$ [4]. The results of large-scale hybrid functional calculations including up to 240 atoms indicate that Σ3 GBs create gap states, alter the size of the band gap and shift the band edges of the light absorber material.

Chemisorption and diffusion of Se and other impurities on Mo surfaces

In addition to the front contact and the light absorber, the interaction between the Mo back-contact and the growth of a thin $MoSe_2$ layer (see Fig. 2) on the Mo surfaces is the second topic of our research. To provide guidelines for our collaborators working in experimental groups and in industrial facilities, we investigated the chemisorption, diffusion, and reaction of Se, Na, K, and O on the Mo(110) surface. Via DFT calculations, we investigated different adsorbate patterns and their scanning tunneling microscopy (STM) images [5-6]. The most stable patterns corresponding to the coverages of $\theta=0.5$ and $\theta=0.75$ monolayers are sketched in Fig. 3, which shows their stability domain.

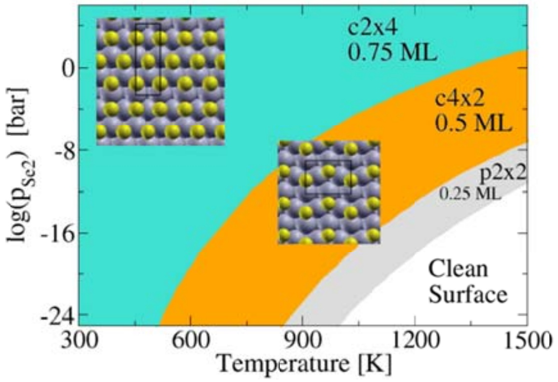

Fig. 3: The stability ranges of the most stable adsorption patterns/coverages of Se on the Mo(110) surface in equilibrium with a gas of Se_2 molecules. The black boxes indicate the surface unit cell.

Regarding the diffusion of Se, Na, and K on the Mo(110) surface, our results from so-called nudged elastic band (NEB) calculations indicate that the presence of Na and K impurities do not change the mass transport of Se atoms on the surface considerably under the conditions of the thin film deposition. In order to provide more accurate predictions of the energetics, morphology, and band alignment of the interface between the Mo back-contact and the $MoSe_2$ layer, we carried out DFT investigations, where the weak van der Waals forces acting between the $MoSe_2$ sheets and between the Mo and $MoSe_2$ were explicitly included in the functional. Using this methodology, we assessed which functionals are able to describe the atomic and electronic structure of the bulk hexagonal $MoSe_2$ and the low-index faces of $MoSe_2$ [7].

We would like to gratefully acknowledge the financial support from the Deutsche Bundesministerium für Wirtschaft und Energie through the ComCIGS II project (0325448C).

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