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Boosting the Open Circuit Voltage of $\text{Cu}_2\text{ZnSnS}_4$ Solar Cells by a Lattice-Matched CeO_2 Layer and Theoretical Understanding of Interface Defects

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The open circuit voltage of state-of-the-art $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$ solar cells with a low S content appears to be limited by bulk recombination. Therefore, at the current stage of development, the standard CdS buffer layer of $\text{Cu}(\text{In},\text{Ga})\text{Se}_2$ solar cells is sufficient for obtaining a high-quality heterointerface with $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$. Conversely, pure-sulfide $\text{Cu}_2\text{ZnSnS}_4$ solar cells are plagued by interface recombination, possibly due to a combination of an unfavorable band alignment with CdS, a large lattice mismatch with CdS, and unfavorable properties of the $\text{Cu}_2\text{ZnSnS}_4$ surface. As a consequence, the open-circuit voltage deficit and efficiency are consistently inferior in $\text{Cu}_2\text{ZnSnS}_4$ solar cells than in $\text{Cu}_2\text{ZnSnSe}_4$ solar cells.

In this work, we tackle the interface recombination problem of $\text{Cu}_2\text{ZnSnS}_4$ solar cells by a combination of theory and experiment. First, we verify by density functional theory that detrimental states within the band gap are expected on $\text{Cu}_2\text{ZnSnS}_4$ surfaces, but not necessarily on $\text{Cu}_2\text{ZnSnSe}_4$ surfaces. This means that band alignment is not the only important parameter when searching for a new buffer material for $\text{Cu}_2\text{ZnSnS}_4$ solar cells. Instead, an ideal buffer material should also remove the native defect states from the $\text{Cu}_2\text{ZnSnS}_4$ surface (passivation role) and form a high-quality interface with $\text{Cu}_2\text{ZnSnS}_4$ without introducing new defect states (by lattice matching and epitaxial growth).

To solve those problems, we propose and test experimentally CeO_2 as a novel buffer layer material in $\text{Cu}_2\text{ZnSnS}_4$ solar cells. The major advantage of CeO_2 is its nearly perfect lattice match with $\text{Cu}_2\text{ZnSnS}_4$ (0.4%), in contrast to the poor lattice match of CdS (7%). CeO_2 is a non-toxic compound that is already used in the fields of catalysis and solid oxide fuel cells. Ce is more earth-abundant than Sn and about as earth-abundant as Cu and Zn (source: U.S. Geological Survey Fact Sheet 087-02).

Here we demonstrate that CeO_2 can be easily grown on $\text{Cu}_2\text{ZnSnS}_4$ by chemical bath deposition. For growth temperatures as low as 50°C , we observe epitaxial growth of CeO_2 on $\text{Cu}_2\text{ZnSnS}_4$ by transmission electron microscopy. Furthermore, CeO_2 films show good surface coverage, low spurious phase content, and a nearly optimal band alignment with $\text{Cu}_2\text{ZnSnS}_4$ as measured by x-ray photoemission spectroscopy.

We then make a first attempt to include CeO_2 in the $\text{Cu}_2\text{ZnSnS}_4$ solar cell architecture by inserting a thin CeO_2 layer between $\text{Cu}_2\text{ZnSnS}_4$ and the usual CdS buffer. The result (over four solar cell batches up to 7% efficiency) is a reproducible open circuit voltage boost compared to the baseline case with a pure CdS buffer layer. The efficiency is also improved in three batches out of four. We argue that this is due to formation of a less defective heterointerface with a lower recombination velocity. By examining the calculated band structure of CeO_2 , we discuss why CeO_2 can be an excellent thin passivation layer for $\text{Cu}_2\text{ZnSnS}_4$ but cannot be used a stand-alone buffer layer.