Searching for a new generation of non-toxic solar cells

Opto-electronic properties of mixed lead-tin metal halide perovskites

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In Short

- Replacement of lead in the metal halide perovskite $CsPbI_3$
- *Ab initio* study of the influence of lead replacement on opto-electronic properties
- Combination of DFT calculation and many-body perturbation theory calculation

Bulk metal halide perovskites have emerged in recent years as solar cell materials with tremendous power conversion efficiency (PCE), reaching those of single-crystal silicon. While the PCE achieved in the laboratory is encouraging, the stability of the samples remains problematic, with samples degrading due to the exposure to air and moisture as well as due to light irradiation. Furthermore, the material contains toxic lead, which may leak from the samples due to degradation. As such, the search for lead-free metal halide perovskites with desirable band gaps, electronic structure, and absorption spectra is ongoing. Mixed metal halide perovksite compounds [1, 2], *i.e.* perovskites where lead has been partially or completely replaced by other metals, particularly tin, have attracted special interest. In this project, we aim to predict accurately the opto-electronic properties of novel compounds by means of *ab initio* calculations, providing important guidelines for further material development. Specifically, we determine first the opto-electronic properties of the lead halide perovskite $\mathrm{CsPbI_3}$ from first principles, and then study how these properties change by increasing the ratio of Sn in $\mathrm{CsPb_{1-x}Sn_xI_3}$ for the values $x = \{1, 0.25, 0.50, 0.75, 1\}$. The schematic structure of one of these compositions, $CsPb_{0.75}Sn_{0.25}I_3$, is shown in Fig. 1.

For each composition, we determine the equilibrium structure from all-electron full-potential density functional theory (DFT) calculations as implemented in the `exciting` code [3]. For these structures, we then determine the electronic structure. We employ the hybrid functional HSE06 for electronic structure calculations. Due to the presence of heavy atoms, such as lead, the effect of spin-orbit coupling on the electronic structure is significant, and is therefore included in the calculations. Finally, optical properties are calculated within many-body perturbation theory (MBPT) through the solution of the Bethe-Salpeter equation (BSE). These calculations provide accurate *ab initio* optical spectra including the effects of electron-hole interaction. As such, we also obtain excitonic binding energies and excitonic wavefunctions.

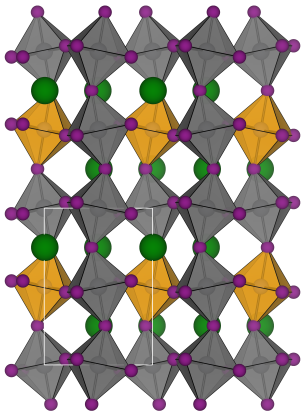

Figure 1: *Schematic structure of $CsPb_{0.75}Sn_{0.25}I_3$ in the orthorhombic phase. Cs atoms are shown in green, iodide ones in purple. The octahedra surrounding lead atoms are shaded in grey, the ones surrounding tin atoms in yellow. The unit cell is indicated by white lines.*

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More Information

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[3] A. Gulans, S. Kontur, C. Meisenbichler, D. Nabok, P. Pavone, S. Rigamonti, S. Sagmeister, U. Werner, C. Draxl, S. Grimme, *J. Phys. Condens. Matter.* **26**, 363202 (2014).

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