**First-principles Calculations of Intrinsic Defect Properties in Halide Double Perovskites for Optoelectronic Applications**

Tianshu Li,1 Mao-Hua Du,2 and Lijun Zhang1,\*

*1School of Materials Science and Engineering, Jilin University, Changchun 130012, China*

*2Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States*

\*E-mail for correspondence: [lijun\_zhang@jlu.edu.cn](mailto:lijun_zhang@jlu.edu.cn)

Lead-free halide double perovskites (HDPs) with the formula of quaternary have recently attracted intense interest as alternatives to lead-halide-perovskite-based optoelectronic materials for their non-toxicity and enhanced chemical and thermodynamic stability. In particular, a series of materials represented by Cs2AgInCl6 and Cs2AgBiX6 (X=Cl, Br) have been successfully synthesized in experiment. With the measured physical properties, including band gaps (2.0-3.0 eV), ultra-long carrier lifetime (6µs for Cs2AgInCl6) and low trap density (towards 108 cm-3), these HDPs have shown desirable potential as useful optoelectronic materials. Controlling defect properties is of vital importance for intrinsic carrier density and transport. Therefore, a comprehensive understanding of carrier trapping at defects and carrier compensation is crucial for the development of HDPs. In this paper, using advanced first-principle energetics calculations, we identify deep-level defects that are detrimental to carrier transport in Cs2AgInCl6, Cs2AgBiCl6 and Cs2AgBiBr6. By evaluating defect formation energies of intrinsic defects using different chemical potentials of composed elements, our calculations reveal the ideal growth conditions to grow *p*-type Cs2AgInCl6 and Cs2AgBiCl6. So they can be potentially used as *p*-type solar absorbers and photon detectors. In contrast, semi-insulating Cs2AgBiBr6 with low carrier density, which is consistent with high observed resistivity in experiment, is a promising semiconductor radiation detection material. Our work provides valuable guidelines for further exploration of Pb-free perovskites for diverse applications.[1]

**References:**

[1] T. Li, X. Zhao, D. Yang, M.-H. Du, and L. Zhang, Phys. Rev. Applied (2018, in press).