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First-Principles Investigation of Ionic Contributions to the Frequency-Dependent Dielectric Function in Cs_2HfCl_6, Cs_2ZrCl_6, and $Cs_2Hf_{0.5}Zr_{0.5}Cl_6$

We present a first-principles investigation of the frequency-dependent dielectric properties of halide double perovskites Cs_2HfCl_6 (CHC), Cs_2ZrCl_6 (CZC), and the mixed alloy $Cs_2Hf_{0.5}Zr_{0.5}Cl_6$ (CHZC). Using density functional theory (DFT) and density functional perturbation theory (DFPT), we evaluated both the electronic ($\varepsilon\infty$) and ionic (ε_ionic) contributions to the dielectric response. While $\varepsilon\infty$ remains relatively consistent across all compositions—indicating similar electronic polarizability—ε_ionic increases from CHC to CZC, due to the more polarizable nature of Zr–Cl bonds compared to Hf–Cl. CHZC exhibits intermediate dielectric behavior, suggesting a smooth compositional transition in lattice polarizability.

Phonon dispersion calculations were performed to confirm the dynamic stability of the materials. Lattice parameters and electronic band structures were calculated within the GGA-PBE functional. Preliminary work is underway to estimate the exciton–phonon coupling strength using the Huang-Rhys factor, while planned GW and GW+BSE simulations aim to provide more accurate exciton binding energies and resolve absorption edge characteristics across the alloy series.

These findings highlight the critical role of ionic lattice dynamics and cation substitution in tuning the dielectric and optical responses, with direct implications for enhancing light yield, scintillation decay characteristics, and energy resolution in radiation detection technologies.

Topical Area

Hard matter: quantum, electronic, semiconducting materials

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