Engineering Transport and Thermoelectric Properties of $Cs_2NaYbCl_6$ Perovskite via Doping and Nanoengineering

Antonio Cappai[1], Claudio Melis[1], Luciano Colombo[1]

1 Department of Physics, University of Cagliari,S.P. Monserrato–Sestu Km 0.700,09042 Monserrato (Cagliari), Italy

(corresponding author: antonio.cappai@unica.it)

ABSTRACT

We present a first-principles investigation of the combined effects of chemical doping and nanostructuring on the thermoelectric performance of the lead-free double halide perovskite $Cs_2NaYbCl_6$. Density functional theory and Boltzmann transport calculations are used to evaluate electrical and thermal transport while explicitly accounting for the main scattering channels: electron-phonon, phonon-phonon, Coulomb impurity, phonon-impurity, and grain-boundary scattering. The calculations show that Coulomb scattering from dopants is efficiently screened and remains negligible compared with the dominant electron-phonon interaction. Consequently, both n- and p-type doping enhance electrical conductivity with only a moderate reduction of the Seebeck coefficient, resulting in a marked increase of the power factor. Phonon-impurity scattering is also weak, whereas nanoscale grain boundaries substantially suppress the lattice thermal conductivity while preserving carrier mobility. By combining optimal n-type doping at 10^{19} cm^{-3} with 10 nm grains, the figure of merit ZT increases from approximately 10^{-8} in the pristine crystal to about 0.12. These results identify controlled extrinsic modification - through doping and grain-size engineering - as a practical route to improve thermoelectric efficiency in wide-bandgap, lead-free halide perovskites.

Keywords: double halide perovskites; thermoelectrics; first-principles calculations; Boltzmann transport; nanostructuring; doping

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