Hessisches Kompetenzzentrum
für Hochleistungsrechnen

High-Throughput Screening of 2D Hybrid Organic-Inorganic Perovskites (HOIPs)



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Clusters
Lichtenberg II Cluster Darmstadt

Software
VASP

Additional Software
Atomic Simulation Environment (ASE), Wannier90

Institute
Theory of Magnetic Materials

University
Technische Universität Darmstadt

Introduction

To meet the ever-growing global energy demand, researchers are continuously exploring new materials. Perovskites represent one such class of materials with the potential to address these energy needs in a sustainable manner. Over the past few decades, scientists have investigated various types of perovskite compounds. Among them, quasi two-dimensional hybrid organic–inorganic perovskites (HOIPs) have emerged as a particularly promising subclass due to their remarkable potential for sustainable energy applications. HOIPs are structurally diverse and contain a large number of atoms. To facilitate the design of new HOIPs, we performed high-throughput first-principles calculations on 549 experimentally known HOIPs, generating approximately 100,000 intermediate structures along with their corresponding energies, forces, and stress tensors for machine-learning interatomic potential (MLIP) training.

Methods

We employed a high-throughput density functional theory (HTP-DFT) workflow to perform a two-step structural relaxations, self-consistent charge-density convergence, and electronic band structure calculations of HOIPs. All calculations used the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) form to describe exchange-correlation effects. We performed k-points convergence before HTP calculations. A kpoints density (*Density = kpoints/2π*) was used for all

structures to ensure converged Brillouin zone sampling. We used a plane-wave energy cutoff of 550 eV for all calculations. For band gap evaluations, we employed both the generalized gradient approximation (GGA) by PBE and DFT-1/2 approaches, with and without spin–orbit coupling (SOC).

Results

We developed a comprehensive hybrid organic–inorganic perovskites (HOIPs) database comprising 100,000 frames, with energies, forces, and stresses calculated at the PBE level. The electronic band structures of experimentally known HOIPs were computed using both the PBE and DFT-1/2 approximations. We observed that both methods fail to accurately describe the band structure of certain HOIPs containing transition metals and heavy elements such as Pb and Bi. In these cases, including spin–orbit coupling (SOC) opens a finite band gap for compounds that were otherwise predicted to be metallic without SOC. We expect that the inclusion of on-site Coulomb interactions within the DFT+U framework will improve the accuracy of band gap predictions for HOIPs containing transition metals. We tested various structure optimization algorithms implemented in the Vienna Ab initio Simulation Package (VASP) and the Atomic Simulation Environment (ASE). Interestingly, some optimizers in ASE, such as BFGS and GPMin, required significantly fewer ionic iterations to reach the force convergence threshold. GPMin, a local Bayesian optimizer, required only 45 iterations, whereas BFGS took 85 iterations and the VASP conjugate gradient algorithm required 145 iterations for a test HOIP containing over 100 atoms.

Discussion

This study establishes a robust data-driven framework that combines high-throughput DFT calculations with machine learning for the design of HOIPs, targeting sustainable applications such as bulk photovoltaics, thermoelectrics, and water splitting. So far, we have generated a database sufficient for training machine learning interatomic potentials on top of a foundation model. In addition, we maintain a database of approximately 500 experimental compounds with their electronic structures calculated using PBE and DFT-1/2, both with and without spin-orbit coupling (SOC). Some compounds could not converge and require higher-level DFT calculations. We are currently performing shift-current calculations for the well converged experimental compounds.

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