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Multi-Fidelity Machine Learning for Perovskite Discovery

ABSTRACT

The ABX3 perovskite crystal structure is ubiquitous and the subject of extensive study owing to the sheer tunability of electronic and optical properties that can be achieved. Halide perovskites, in particular, are materials of great interest for solar absorption and many related optoelectronic applications such as LEDs, lasers, and UV or IR sensors. The discovery of novel perovskite compositions, including complex alloys with attractive properties, is hindered by the combinatorial nature of the chemical space and a general lack of quantification of systematic inaccuracies in simulations such as from first principles-based density functional theory (DFT). In this work, we generated large datasets of computed stability, electronic band gaps, theoretical photovoltaic efficiency derived from optical absorption spectra, and defect formation energies, of halide perovskite alloys from various DFT semi-local and hybrid functionals. This data is combined with smaller quantities of corresponding experimental measurements collected from the literature, and used for training multi-fidelity machine learning (ML) models to make property predictions at experimental accuracy. Such predictions, using state-of-the-art nonlinear regression techniques including random forests and Gaussian processes, are sequentially improved and coupled with a recommendation engine for new computations and experiments to gradually achieve new stable compositions with targeted band gap and absorption.

While we obtain high prediction accuracy using only compositional information and known elemental properties of A, B, and X species as inputs, we further explore the possibilities of using entire crystal structures as input via a variety of graph neural network-based approaches. The latter approach enables inclusion of lattice strain, octahedral distortions and rotations, and different kinds of alloy ordering as implicit inputs to the ML framework, ultimately resulting in general models applicable to any atom-composition-structure combination within the selected halide perovskite chemical space. Best DFT-ML surrogate models are coupled with optimization schemes using methods such as genetic algorithm, used to drive collaborative experiments and further DFT computations, and made available to the community via user-friendly tools on the NSF-funded online repository, nanoHUB.

BIO

Arun Mannodi Kanakkithodi is an Assistant Professor in the School of Materials Engineering at Purdue University. He received his PhD in Materials Science and Engineering from the University of Connecticut in 2017, working in Prof. Rampi Ramprasad's research group. Arun worked as a postdoctoral researcher in the Nanoscience and Technology Division at Argonne National Laboratory from 2017 to 2020, working with Dr. Maria Chan. His research focuses on applying high-throughput density functional theory (DFT) and machine learning (ML) for accelerating the discovery of novel materials for a variety of energy applications. Currently, the Mannodi research group at Purdue works on using DFT-ML for studying point defects and dopants in semiconductors, for structure-composition engineering of halide perovskites, and studying chalcogenide semiconductors for photocatalysis. Arun received the Distinguished Young Investigator award in 2020 and the Outstanding Postdoctoral Performance award in 2019 at Argonne. He is a regular organizer of monthly hands-on ML workshops with nanoHUB and the lead coordinator of "intro to ML" workshops at the Materials Research Society (MRS) fall and spring meetings as member of the MRS AI Staging Committee.

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