Finite-temperature band gaps at scale with multi-fidelity foundation machine learning interatomic potentials

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The fundamental band gap is a critical parameter dictating the performance of semiconductors, optoelectronics, and energy-conversion devices. Most computationally evaluated band gaps are reported at 0 K, yet devices operate at room temperature or above. Thus, accurately predicting band gaps at finite temperatures remains a formidable challenge. Standard density functional theory functionals severely underestimate experimental values, while highly accurate hybrid functionals are computationally prohibitive for the extensive structural sampling required at finite temperatures. To overcome this limitation, we developed MACE-bg, a modification of the MACE interatomic potential [2] that can provide highly accurate band-gap evaluations trained alongside potential energy surfaces (energy, forces, stresses). Using MACE-bg, we trained `MACE-bg-MH1-h4`, a novel multi-head machine learning model based on the MACE interatomic potential architecture, fine-tuned from the foundational MACE-MH1 model [1]. Our architecture enables band-gap prediction across multiple fidelity levels: PBE, r^{2}SCAN, HSE06, and direct experimental data. By training on these diverse fidelities, the model reaches the accuracy of state-of-the-art machine learning band gap models, while preserving excellent prediction accuracy for energies, forces, and stresses trained on two of these fidelities (PBE and r^{2}SCAN), with the efficiency of MACE-MH1 models. Crucially, this multi-fidelity approach lets researchers select their desired level(s) of theoretical or experimental accuracy while naturally accounting for finite-temperature structural dynamics through molecular dynamics. Furthermore, we compute finite-temperature band gaps for 41,230 materials from the Materials Project database [3], a scale previously impossible at hybrid and experimental accuracy. We find that 14.8% of materials widen their gap upon heating, contrary to the usual behavior [4, 5]. The effect is especially visible in compounds containing d^{10} coinage metals and ns^2 lone-pair groups, reproducing families known from experiment, such as cuprous halides, halide perovskites, and lead chalcogenides, and we identify four further classes with no previous literature reports, including coinage-metal fluorides and Cr(IV) in oxides and fluorides.

References

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