Predicting properties of 2D materials using graph neural network

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In recent years, data-driven techniques in materials science have garnered attention for their adeptness in – efficiently exploring the vast landscape of material properties, generating novel structures and assisting traditional simulation methods with machine-learned interatomic potentials (MLIPs). Particularly, generative methods have emerged as powerful tools for structure exploration, enabling the discovery of novel materials with tailored functionalities.
In previous work [1], it was shown that a crystal diffusion variational autoencoder (CDVAE) can generate two-dimensional (2D) materials of high chemical and structural diversity and formation energies mirroring the training structures. The CDVAE was trained on 2615 2D materials with energy above the convex hull ΔH_{hull} < 0.3 eV/atom, and generated 5003 structures, which were then relaxed using Density Functional Theory (DFT). Along with CDVAE, lattice decoration method was used to generate 14192 new crystals by systematic element substitution of the training structures. It was found that the generative model and lattice decoration approach are complementary and yield materials with similar stability properties but very different crystal structures and chemical compositions. In total, 11630 new 2D materials were predicted, out of which 3331 are within 100 meV of the convex hull and could potentially be synthesised.

While generating structures is crucial, calculating their properties with ab-initio methods remains a challenging endeavour, often requiring extensive computational resources. For example, an extensive DFT study was done on the 3331 most stable 2D materials found from the generative model, assessing the thermodynamic stability and chemical validity of the structures. About 70% (2759 structures) of them are predicted to be dynamically stable, on which the C2DB workflow was employed to compute various material properties like - the stiffness tensor, piezoelectric tensor, deformation potentials, Born and Bader charges, electronic band structure, effective masses, plasma frequency, Fermi surface, projected density of states, magnetic moments, magnetic exchange couplings, magnetic anisotropy, topological indices, optical- and infrared polarisability. This work [1, 2] exposes previously unknown parts of the 2D chemical space and provides a basis for the discovery of 2D materials with specific properties. All data is available in the C2DB.

Finding new stable crystals and their characterisation of the basic material properties is crucial for bridging the gap between computational materials discovery and their synthesis for experiments and applications. The progress in generative models has given rise to vast amounts of new crystal structures, to the point where using ab-initio methods directly can be challenging.
In this work, we will employ graph neural networks to predict some of the properties from the C2DB workflow, acting as a screening step before a high-throughput study, thereby accelerating discovery of promising generated materials. This substantial 2D materials database serves as an extensive training set for the development of a novel C2DB-ML database. Leveraging the CDVAE and lattice decoration protocol, we generate thousands of additional 2D structures, which are then relaxed using machine learning (ML) universal interatomic potentials, fine-tuned based on nearly 12,000 DFT-relaxation trajectories from our initial generated materials. Moreover, we will employ graph neural network, trained on C2DB to predict, instead of calculating, various properties mentioned in the C2DB workflow. We envision this comprehensive ML database to act as a foundation for high-throughput screening studies, facilitating the identification of promising candidate materials for further exploration using the ab initio DFT workflow on a select subset of intriguing candidates.

References

[1] P Lyngby, KS Thygesen, npj Computational Materials 8 (2022) 232
[2] P Lyngby, KS Thygesen, (2024) arXiv:2402.02783
[3] T Xie, (2021) arXiv:2110.06197

[4] S Feng (2024) arXiv:2403.11347v1