

## Computational design of stacking oxide free-standing membranes into artificial heterostructures

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Oxide materials have shown a wide range of properties attractive for energy device applications. Novel phenomena occur especially at the interface [1], for example 2D electron gas [2], 2D hole gas [3], and ferromagnetism [4]. Previous approaches have utilized epitaxial growth to enable atomic-level control of the interface. However, this limits the interface to material systems with a good lattice match between the film and substrate.

New methods have produced ultra-thin freestanding oxide membranes [5]. In the NEXUS ERC project, we aim to produce artificial heterostructures by stacking these membranes (fig 1.), allowing complete control of the interface, without the limitations of epitaxy.

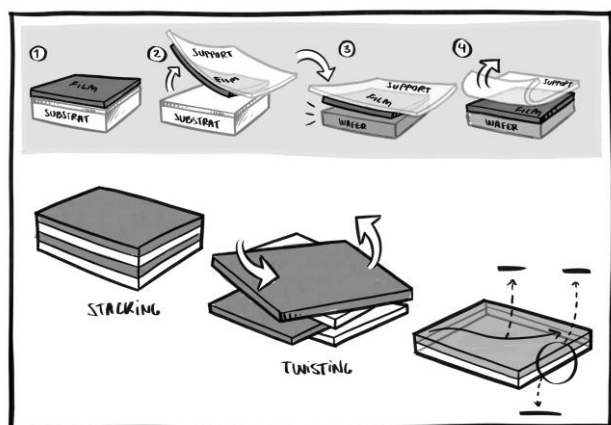


Figure 1. Stacking of thin-film oxide membranes

This methodology opens several degrees of freedom for building the heterostructure, such as the materials stacked and the twist angle of the membranes, providing a way to tune the properties of the materials [6]. By engineering the device, we aim to find fast ionic paths through dislocation networks, potentially breaking the fundamental limits of existing energy devices.

In this pursuit, an in-depth theoretical understanding of the interfacial reconstruction and stability of the artificial heterostructures is required. A theoretical understanding will provide both insight to what enables or hinders fast ionic conductivity and will allow inverse design of the heterostructures to optimize the conductivity.

The complex heterostructures that can be engineered may require several thousand atoms to

describe theoretically, due to the Moiré patterns that occur when twisting, making it nearly impossible to describe with density functional theory. Therefore, we will use message passing neural networks that have shown excellent accuracy as interatomic potentials, when trained on a high-quality data set [7].

Furthermore, it's been shown that traditional generalized gradient approximation functionals do not accurately describe the complex electronic structure in transition metal oxides [8], thereby not accurately capturing the transition states involved in the ionic diffusion. To alleviate this issue, we will construct a data set of hybrid-level calculations.

The sizable task of creating a data set of hybrid-level calculations sufficient for training an accurate machine learning interatomic potential is made possible by employing the batch active learning workflow CURATOR [9]. By utilizing molecular dynamics exploration and active learning, CURATOR efficiently explores the potential energy surface, reducing the computational cost of data generation (fig 2.).

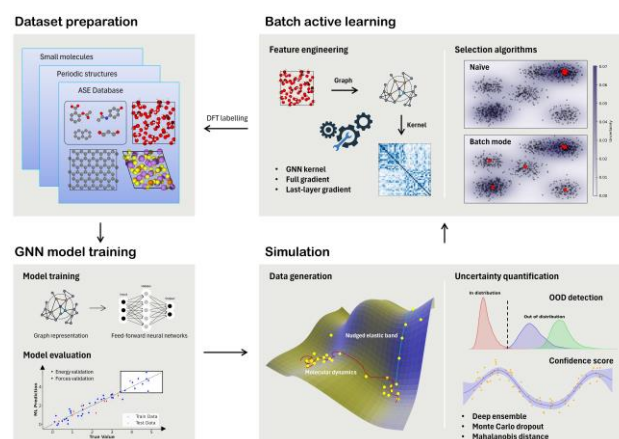


Figure 2. Data generation workflow [9]

Furthermore, we aim to go beyond state-of-the-art MLIPs and additionally train new models such as DeepDFT [10] that can predict the electronic structure. Thereby, we will significantly enhance the theoretical knowledge elicited from predictions of the model, to better explain the complex correlation phenomena occurring in the transition metal oxide. This is crucial, as the interesting properties of transition metal oxides occur in large part due to the mixed ionic-covalent nature of the transition metal and oxide bonds.

The value of an accurate machine learning interatomic potential based on hybrid-level DFT is not limited to engineering fast ionic oxide heterostructures. In the rapidly developing field of freestanding oxide membranes, a wide range of properties can prove groundbreaking. For example, novel ferroic properties can prove integral for oxide electronics [11]. The MLIP can provide valuable theoretical insight to understand these phenomena as the field develops.

## References

- [1] The interface is still the device. *Nature Mater* **11**, 91 (2012)
- [2] Santander-Syro, A., Copie, O., Kondo, T. et al. *Nature* **469** (2011) 189–193
- [3] Lee, H., Campbell, N., Lee, J. et al. *Nature Mater* **17** (2018) 231–236
- [4] Brinkman, A., Huijben, M., van Zalk, M. et al. *Nature Mater* **6** (2007) 493–496
- [5] Lu, D., Baek, D., Hong, S. et al. *Nature Mater* **15** (2016) 1255–1260.
- [6] Li, Y., Pryds, N., et al. *Adv. Mater.* **34** (2022) 2203187
- [7] Batzner, S., Musaelian, A., Sun, L. et al. *Nat Commun* **13** (2022) 2453
- [8] Ricca, Chiara. et al. *Phys. Rev. Res.* **2** (2020) 023313
- [9] Yang X, et al. *ChemRxiv* (2024)
- [10] Jørgensen, P.B., Bhowmik, A. *npj Comput Mater* **8** (2022) 183
- [11] L. Han, G. Dong, M. Liu, Y. Nie, *Adv. Funct. Mater.* **34** (2024) 2309543