

Machine Learning Accelerated Geometry Optimization under External Electric Fields

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Abstract—This work introduces a hybrid machine learning (ML) and density functional tight-binding (DFTB) framework to accelerate geometry optimization under external electric fields. The method combines a machine learning potential (MLP) for the (usual) Hellmann–Feynman (HF) force with a computationally inexpensive evaluation of the field-induced ionic force, obtained from predicted Born effective charges (BEC) or DFTB calculations. Validation in an H₂O molecule demonstrates reorientation of the dipole moment, and the H-O-H angle is in good agreement with density functional theory (DFT). We further apply the framework to SiO₂ and HfO₂ slabs to capture the ionic contribution to permittivity. The resulting static permittivity values agree well with reported values. The framework is scalable to large systems, paving the way for efficient calculations of electromechanical properties in nanoscale systems.

I. INTRODUCTION

Geometry optimization under external electric fields is essential in modeling semiconductor devices, as it enables accurate predictions of material properties and device performance under realistic operating conditions. DFT is accurate but computationally expensive, limiting simulations to small systems and hindering the study of realistic phenomena such as interfaces, defects and amorphous structures. MLPs have emerged as an efficient alternative with near-DFT accuracy for energies and forces predictions. However, incorporating external electric fields is challenging, and coupling to arbitrary electrostatic boundary conditions remains unexplored. This work addresses these gaps by: 1) Predict the HF force using an MLP, 2) Obtain the field-induced ionic force using either predicted BEC or DFTB calculations.

II. METHODOLOGY

The first-principles determined total force under an external electric field is [1]:

$$F_{\text{tot}} = F_{\text{HF}} + F_{\text{EXT}}, \quad (1)$$

where F_{HF} and F_{EXT} are the HF force and the field-induced ionic force, respectively. The HF force is inferred from an MLP trained on DFT data without external electric fields.

For F_{EXT} , we considered two approaches. Firstly, we write:

$$F_{\text{EXT}} = Z^* E, \quad (2)$$

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where Z^* is the BEC. The BEC tensors are predicted using an in-house equivariant neural network (the architecture is shown in Fig.1) trained on DFT data. Equivariant neural networks have built-in rotational equivariance, suitable for predicting vectorial and tensorial quantities [2].

To generate the necessary training data, we compute the first-principles Born effective charges in several different ways, using our in-house density functional theory code and Quantum ESPRESSO. We use the PBE exchange–correlation functional and norm-conserving pseudopotentials throughout. To obtain the most accurate reference values, we use plane-wave density functional perturbation theory (DFPT) and compute the Born effective charges from the induced phonon response and the bare electric perturbation:

$$Z_{\alpha\beta}^* \propto \langle \psi_\alpha | v_\beta^E | \psi_0 \rangle. \quad (3)$$

Here, ψ_α is the linear-response wave function to the phonon perturbation, and v_β^E is the potential corresponding to the external electric field, whose action on the ground-state wave functions ψ_0 is computed using the commutation relation. This method is applicable to bulk systems and to systems of at most a few hundred atoms. Beyond this size regime, the plane-wave DFPT approach becomes too expensive and is therefore challenging to apply to interfaces. In that case, we instead adopt a finite electric field approach. Here, a series of small clusters/nanocrystals is cut out from a prototypical Si/SiO₂ interface and the surface dangling bonds are hydrogenated. A finite positive and negative sawtooth field is then applied, with a magnitude small enough that the induced response remains in the linear regime. Overall, this method requires only six self-consistent calculations (two for each Cartesian direction). The Born effective charge is extracted from the change in force due to the applied external electric field (Fig.2):

$$Z_{\alpha\beta}^* = -\frac{\partial F_\alpha}{\partial E_\beta}. \quad (4)$$

Alternatively, we compute the field-induced force using DFTB:

$$F_{\text{EXT}} = F_{\text{DFTB1}} - F_{\text{DFTB0}}, \quad (5)$$

where F_{DFTB1} and F_{DFTB0} are the forces calculated with and without the external electric field, respectively. Atomic forces in DFTB are obtained by taking gradients of the total

energy, which includes contributions from the band structure, Coulomb interactions, and repulsive potentials [3]. Since the electronic response to the external electric field naturally merges from the DFTB Hamiltonian when coupled to Poisson's equation [4], the field-induced force can be calculated by taking the difference of DFTB forces with and without the external electric field. Because the repulsive term depends only on atomic positions, it cancels out and only the band structure and Coulomb contributions are relevant for the field-induced force. Although the field-induced force calculated using BEC directly quantifies the ionic contribution, the DFTB-based approach avoids the need to train a separate ML model for BEC, and can be extended to systems where the concept of BEC may not be well-defined, such as metal-insulator-semiconductor interfaces. Furthermore, when coupled with a Poisson solver, it enables simulations under arbitrary electrostatic boundary conditions.

III. RESULTS

Starting from a water molecule in equilibrium with the dipole oriented along the $-z$ direction, we relax it under an applied external electric field along $+x$. The optimized geometry exhibits dipole reorientation (Fig.3) and a relaxed H–O–H angle consistent with DFT calculations (Table I).

We then apply the method to SiO_2 and HfO_2 slabs. The training results and the DFTB calibrations are shown in Fig.6. After relaxing the structure with and without the external electric field, we extract the low-frequency permittivity profile using the continuity of the electric displacement field [4]. This translates to $\epsilon_i E_i = \epsilon_{i+1} E_{i+1}$, where ϵ and E are the permittivity and the induced field, and i and $i+1$ indicate two neighboring layers ($i=0$ is in the vacuum region where $\epsilon_i = 1$). The static permittivity values obtained for both materials (Fig.9) are consistent with reported values [5], [6].

The local permittivity profile of the SiO_2 slab is shown in Fig.7. The high-frequency (optical) permittivity corresponds to the electronic response at fixed ionic coordinates, whereas the low-frequency (static) permittivity is extracted after the structure is relaxed under the applied electric field. The low-frequency permittivity is larger than the high-frequency value due to the additional ionic contribution to the polarization. The oscillations and deviations near the slab interfaces reflect the finite-thickness geometry and the spatially nonlocal nature of the microscopic dielectric response.

A single A100 GPU is used to test the scalability of the proposed BEC method. For systems with 38, 152, 342, 608 and 2432 atoms, the turnaround time per each relaxation step is 0.8, 1.0, 1.3, 1.6 and 12.8 seconds. For the DFTB method, the turnaround time is decided by the DFTB SCC calculations.

IV. SUMMARY

This work demonstrates a scalable ML and DFTB framework for geometry optimization under electric fields, validated on H_2O molecule, SiO_2 and HfO_2 slab systems.

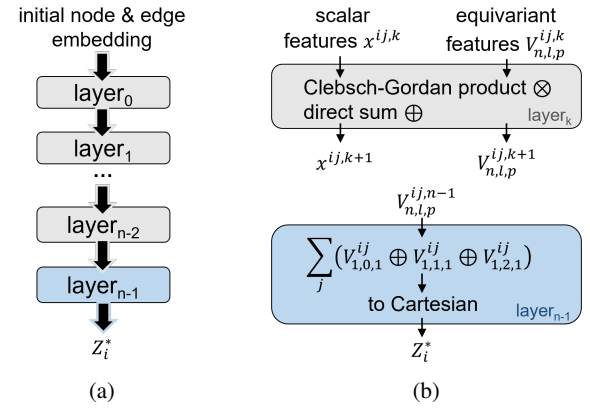


Fig. 1: (a) model architecture for BEC prediction and (b) the interaction and the final layer, where i and j are the atom indices, k is the layer index, n, l and p label the channel, the order and the parity of the spherical harmonics, respectively.

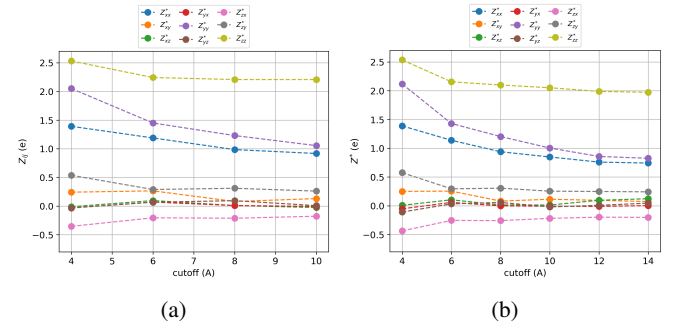


Fig. 2: (a) shows the Born effective charge of the center atom at the interface as a function of the cluster diameter. The computed values plateau and are therefore deemed converged at around 10 Å. As a final step to improve computational efficiency, we also tested a localized-basis approach, in which the wave functions are expanded in strictly localized numerical radial orbitals. For this test, the pseudopotentials and basis functions of OpenMX are used, with a DZP basis [7]. (b) shows close agreement with the plane-wave approach. Notably, these calculations are at least an order of magnitude faster for the same computational resources. We anticipate that, going forward, all calculations will be performed using this localized-basis approach.

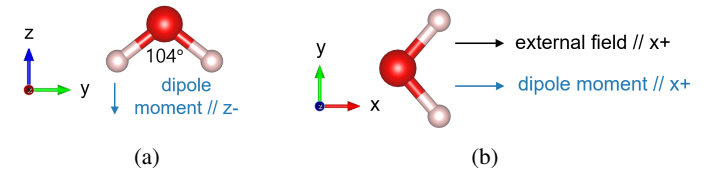
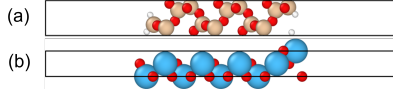
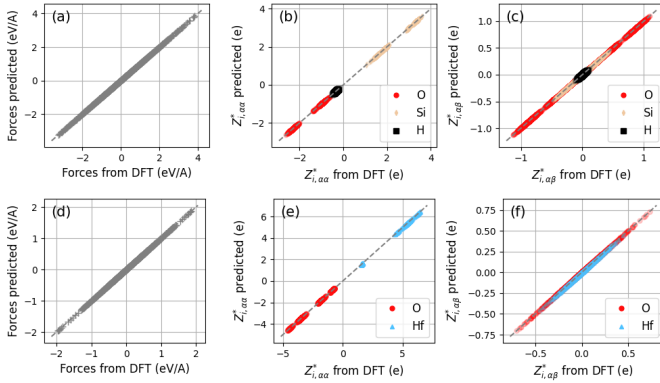
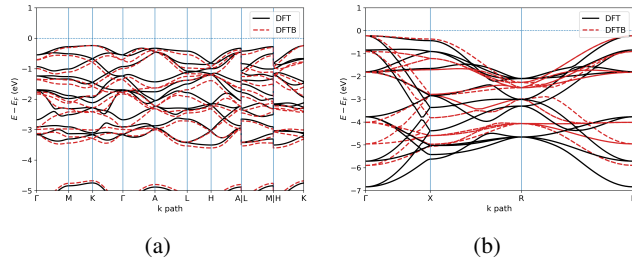
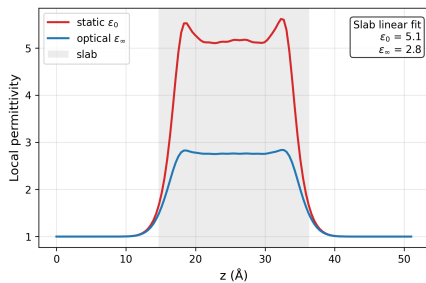
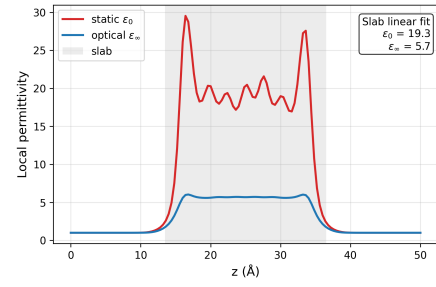
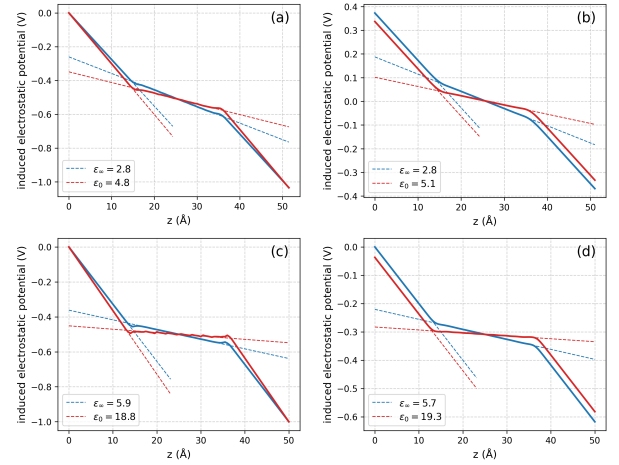


Fig. 3: Initial (a) and relaxed (b) structures of H_2O molecule under external electric field.

TABLE I: H-O-H angles under external electric fields.

External field (V/Å)	DFT (°)	BEC (°)	DFTB (°)
1	102.25	102.28	102.36
1.25	101.57	101.62	101.55
1.5	100.88	100.99	100.67

Fig. 4: SiO₂ and HfO₂ slabs for permittivity calculations.Fig. 5: Training results for SiO₂ (upper) and HfO₂ (lower) slabs. Forces (a and d), diagonal (b and e) and off-diagonal components of BEC (c and f) are shown.Fig. 6: In-house DFTB database, band structure of (a) SiO₂ and (b) HfO₂ can be well reproduced against DFT.Fig. 7: Local permittivity of the SiO₂ slab. The structure is relaxed using the MLP+DFTB method.Fig. 8: Local permittivity of the HfO₂ slab. The structure is relaxed using the MLP+DFTB method.Fig. 9: Linearly fitted permittivity for SiO₂ ((a) & (b)) and HfO₂ ((c) & (d)) slabs, using BEC ((a) & (c)) and DFTB ((b) & (d)) for the field-induced ionic force. For BEC, a constant field of 0.02 V/Å is applied, while for DFTB, Neumann boundary condition of 0.02 V/Å is applied. Slopes of the induced electrostatic potential (dashed lines) in the vacuum and the slab region are used to extract the permittivity. Blue and red curves are induced electrostatic potential w/o and w/ structure relaxation under the external electric field, respectively.

REFERENCES

- [1] H. Fu and L. Bellaiche, "First-principles determination of electromechanical responses of solids under finite electric fields," *Phys. Rev. Lett.*, vol. 91, p. 057601, Jul 2003.
- [2] N. Thomas, T. Smidt, S. Kearnes, L. Yang, L. Li, K. Kohlhoff, and P. Riley, "Tensor field networks: Rotation- and translation-equivariant neural networks for 3d point clouds," 2018.
- [3] P. Koskinen and V. Mäkinen, "Density-functional tight-binding for beginners," *Computational Materials Science*, vol. 47, no. 1, pp. 237–253, 2009.
- [4] S. Markov, G. Penazzi, Y. Kwok, A. Pecchia, B. Aradi, T. Frauenheim, and G. Chen, "Permittivity of oxidized ultra-thin silicon films from atomistic simulations," *IEEE Electron Device Letters*, vol. 36, no. 10, pp. 1076–1078, 2015.
- [5] C. Sevik and C. Bulutay, "Theoretical study of the insulating oxides and nitrides: SiO₂, GeO₂, Al₂O₃, Si₃N₄, and Ge₃N₄," *Journal of Materials Science*, vol. 42, pp. 6555–6565, Aug 2007.
- [6] N. Shi and R. Ramprasad, "Local dielectric permittivity of hfo2 based slabs and stacks: A first principles study," *Applied Physics Letters*, vol. 91, p. 242906, 12 2007.
- [7] T. Ozaki and H. Kino, "Numerical atomic basis orbitals from h to kr," *Phys. Rev. B*, vol. 69, p. 195113, May 2004.

