AI²MD (Artificial Intelligence × Ab Initio Molecular Dynamics) Simulation of Electrochemical Interfaces: Methods and Applications

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WHY we need AI²MD for EC studies

While *ab initio* simulations have provided valuable **microscopic** insights into EC interfaces, the high computational cost limits their use in tackling complex systems. Machine learning potentials offer a solution, but their application in electrochemistry remains challenging due to the difficulty in **treating the dielectric response of electronic conductors and insulators simultaneously**…

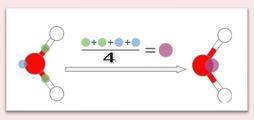
AIMD for EC interfaces

- **BPNN**: Behler and Parrinello, PRL **2007**
- **GAP**: Bartók, Csányi *et al.* PRL **2009**

AI²MD for S/L interfaces

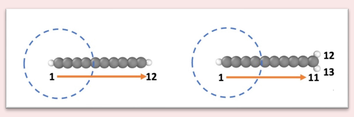
AI²MD for EC interfaces

Electrolyte



Zhang, Car, *et al. JCP* **2022**

✓Reorientational response



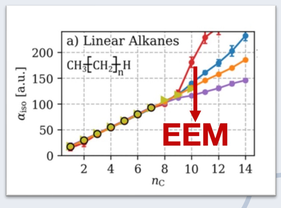
Ko, Behler, *et al. Nat. Commun.* **2021**

✗Non-local elec. dielectric response

Electrode

Ko, Behler, *et al. Nat. Commun.* **2021**

✓Non-local CT



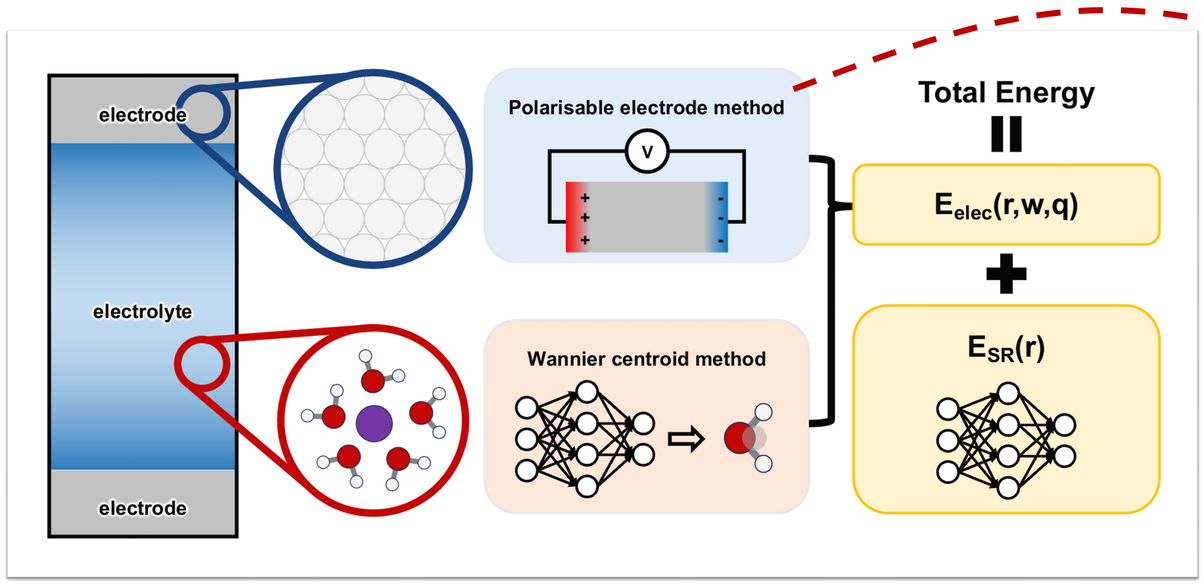
Shao, Zhang, *et al. Electron. Struct.* **2022**

✗Overestimate polarisability

HOW we achieve AI²MD

Therefore, we propose a **hybrid scheme** of MLPs to treat electrochemical interfaces (**ec-MLP**):

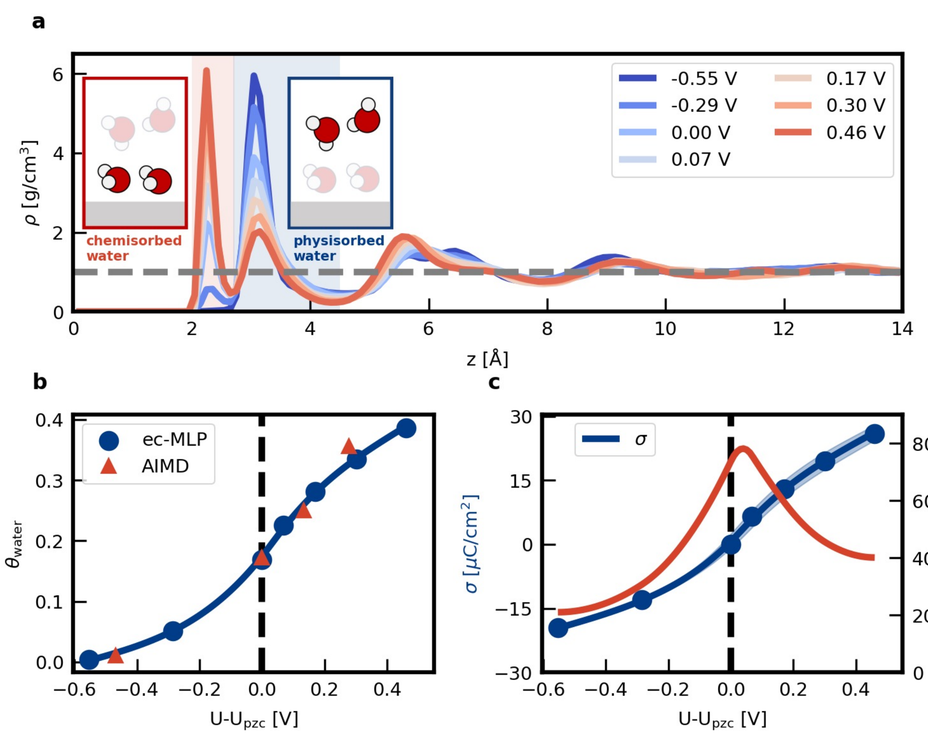
- **Wannier centroid (WC) method** for electrolytes (ionic conductor & electronic insulator)
- **Polarisable electrode method** for electrodes (ionic insulator & electronic conductor)



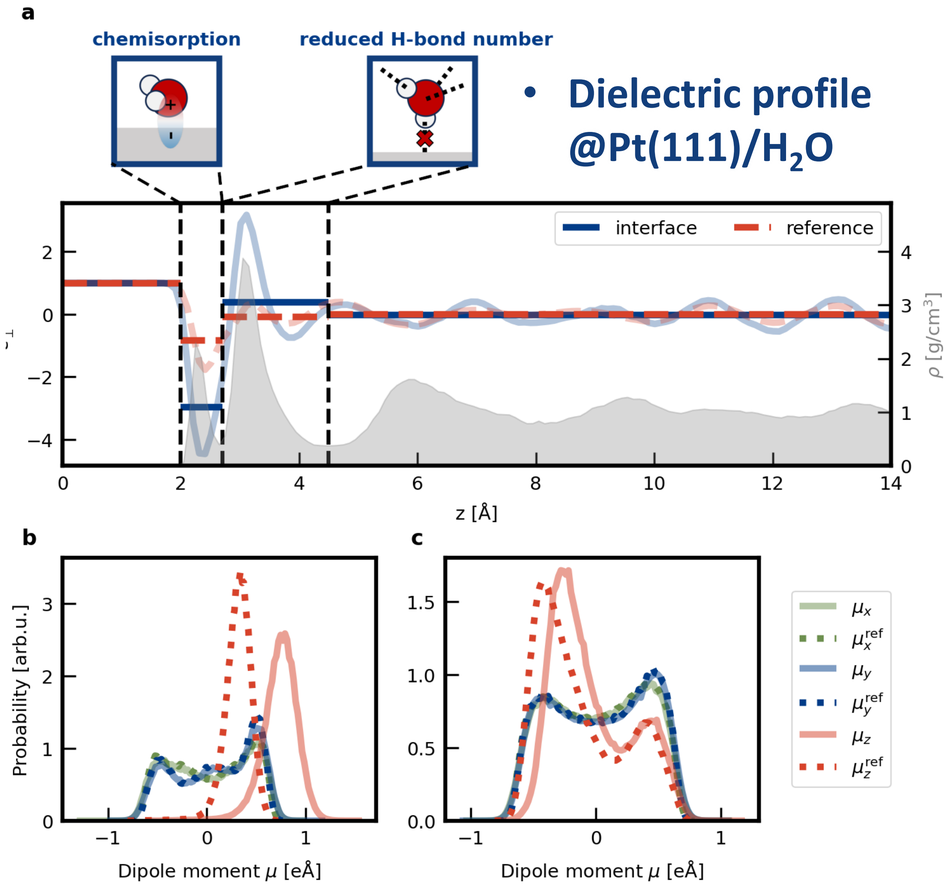
Extendable for more accurate methods, e.g., charge equilibrium (**QEq**) method and orbital-free DFT (**OF-DFT**).

- **Chemical potential**
- **Local environment**

WHAT we can obtain from AI²MD



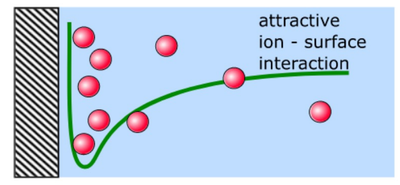
- **Pt(111)/KF(aq) interfaces**
- **Potential-dependent water ad-/de-sorption**
- **Bell-shaped Helmholtz capacitance** (which requires accurate descriptions of electronic structures!)



- **Dielectric profile @Pt(111)/H_2O**
- **Chemisorption induces additional dipoles**
- **Physisorption lower dipole due to lower HB number**

Coming soon…

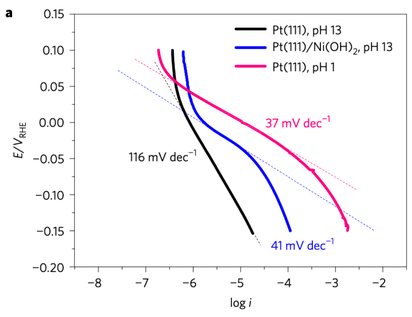
EDL structures



Doblhoff-Dier, Koper, *JPCC* **2021**

Ojha, Doblhoff-Dier, Koper, *PNAS* **2022**

pH/Ion effects



Ledezma-Yanez *et al.*, *Nat. Ener.* **2017**

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