Machine Learning-Driven Adsorption Simulations in RASPA3 using `raspalib` python interface

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Predicting adsorption in metal organic frameworks (MOFs) remains difficult when water or CO_2 interacts with coordinatively unsaturated metal sites: conventional, two-body, force fields can miss these interactions, while rigid-host models neglect adsorption-induced structural response, including framework breathing. We are extending RASPA3 [1] and its Python interface, `raspalib`, to integrate learned models directly into established Monte Carlo and molecular dynamics workflows. Here, we implement foundation machine learning interatomic potentials (MLIP), such as MACE in the RASPA3 simulation routines.

Through `raspalib`, interatomic energies and forces can be supplied by callback functions, enabling machine-learned potentials to act as components of the simulation engine rather than stand-alone calculators. For message-passing models such as MACE [2], a dedicated C++ engine caches graph features between Monte Carlo trials. Displacements, insertions and deletions update only the affected receptive field while preserving the full-system energy, including many-body interactions. The same interface also supports molecular dynamics and volume-changing moves. Initial simulations demonstrate an end-to-end route from a pretrained model to dynamical trajectories and grand canonical adsorption calculations.

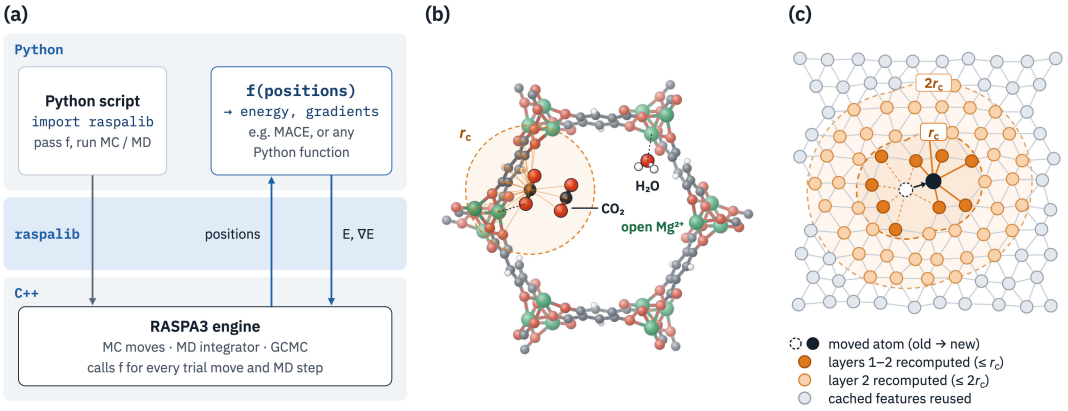

Figure 1: (a) Python runs RASPA3 through `raspalib`. The energy can be any function $f(\mathbf{r}) \rightarrow (E, \nabla E)$, here wrapping MACE, which the C++ engine calls at every trial move and MD step. (b) CO_2 and H_2O at open Mg^{2+} sites in Mg-MOF-74; the dashed sphere marks the cutoff r_c. (c) After a trial move, only features within r_c (layer 1) and $2r_c$ (layer 2) are recomputed. All others are reused from the cache, and the full-system energy is preserved.

Our explicit application targets are water and CO_2 adsorption in Mg-MOF-74 and Cu-BTC (HKUST-1). Both materials present chemically specific metal-adsorbate interactions, providing demanding test cases for evaluating adsorption configurations, loading behaviour and the role of many-body adsorbate-adsorbate interactions.

In parallel, we are connecting neural classical density functional theory [3] to RASPA3. In this complementary description, an equilibrium adsorbate density replaces explicit guest sampling; differentiation with respect to framework coordinates yields adsorption-induced mean forces. This provides a route to coupling adsorption thermodynamics with framework dynamics without tracking individual adsorbate trajectories.

Together, these developments position RASPA3 and `raspalib` as an extensible computational platform for investigating adsorption and, ultimately, adsorption-induced deformation and breathing in flexible porous materials.

References

[1] Y. A. Ran *et al.*, *J. Chem. Phys.* **161**, 114106 (2024).

[2] I. Batatia *et al.*, *Adv. Neural Inf. Process. Syst.* **35**, 11423–11436 (2022).

[3] J. Dijkman *et al.*, *Phys. Rev. Lett.* **134**, 056103 (2025).