Modelling complex, stimuli-induced order–disorder transitions in metal–organic frameworks

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Metal–organic frameworks (MOFs) are a class of porous functional materials that consist of metal nodes linked by organic molecules (Fig. 1a) [2,3]. MOFs demonstrate complex behaviours under changes in temperature, mechanical stress, or guest adsorption [3–7]. These thermodynamic stimuli often induce reversible or irreversible transitions to states that lack long-range or framework order (Fig. 1b).

These disordered states are of interest for functional applications and stand out due to their structural adaptability, *e.g.*, the capacity to dissipate strain without significant material degradation [4]. However, both experimental and computational challenges in analysing these disordered atomic structures leave the transition behaviours of MOFs unpredictable, preventing their rational design and application.

Herein, we address these challenges using an *in silico* approach based on machine-learned (ML) potential models to elucidate and understand the structures of disordered MOFs, and—where possible—understand the mechanisms behind these order-to-disorder transitions [8–10]. Our focus is on deriving models that a sufficiently accurate to distinguish between transitions for different MOF linkers, while also being efficient enough to scale to model sizes for which meaningful comparison with experimental data is possible. To this end, we use the ML potential models to drive atomistic simulations of the disordered MOFs, which we validate against experimental X-ray diffraction and pair distribution data for the disordered MOF forms [3,6,7].

Figures

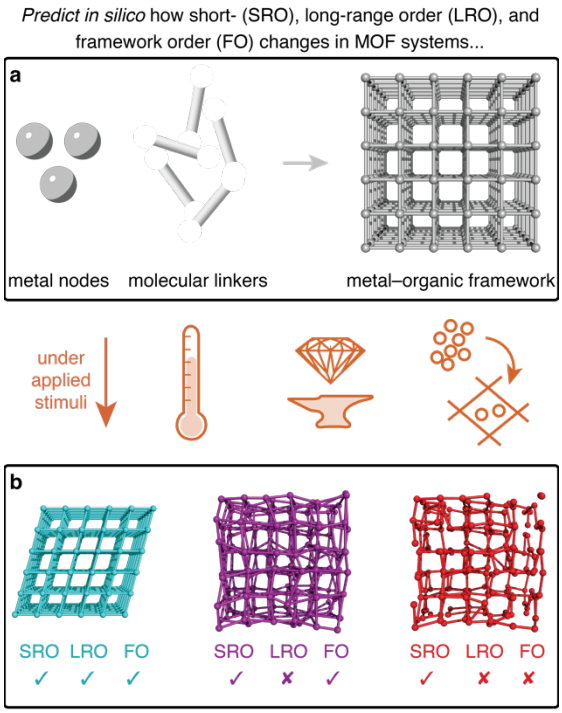

Figure 1. Stimuli-induced transitions in MOFs. a, MOFs are composed of building units that form extended framework structures. Under external stimuli, MOFs transition to various states depending on their constituent building units. **b,** These states are distinguished by the degree of structural order across different length scales.

References

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