Invited Lecture

Investigating amorphous materials with total scattering and coarse-grained approaches

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Total scattering experiments provide valuable information on the local and intermediate-range order in complex materials. By taking the Fourier transform of a total scattering function, you obtain the pair distribution function (PDF): a weighted histogram of interatomic distances. Given a structure model of a material, consisting of the coordinates of the atoms, it is straightforward to compute the PDF and therefore validate the model against experiment. However, generating these structural models from the PDF data—the so called 'inverse problem'—is challenging because meaningfully different structure models can give rise to identical PDFs.

In this contribution, I will describe our approach to generating high-quality amorphous structures by considering the relative energies of the models. Provided there exists an *accurate* description of the potential energy surface for the system of interest, the simultaneous refinement of atomic coordinates to total scattering data while also minimising the total energy leads to greatly improved structural solutions, compared to just fitting the experimental data alone [1].

I will demonstrate this approach for two case studies: amorphous calcium carbonate (ACC) [2] and an amorphous metal–organic framework (a-MOF) [3]. For ACC, we make use of state-of-the-art empirical potentials [4], while for the a-MOF, we develop a machine learned potential [5]. In both cases, our structural solutions, alongside coarse-grained analysis, allow us to develop a deeper understanding of the structure and complexity of these amorphous materials [1].

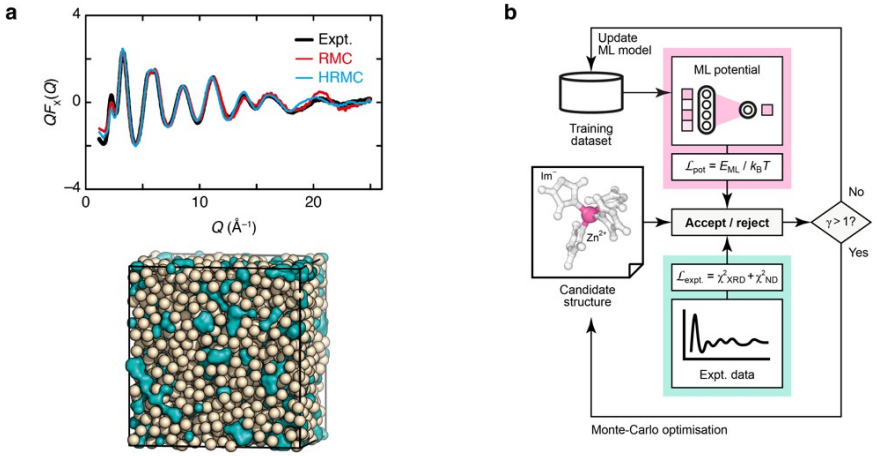

Figure 1. **Improving amorphous modelling by combing total scattering and *accurate* energetic descriptions. a**, The combined approach gives an improved model of amorphous calcium carbonate consistent with X-ray total scattering data and state-of-the-art empirical potentials. **b**, An active learning scheme facilitates the development of a machine learned potential capable of accurately predicting the atomic energies and forces in an amorphous metal–organic framework.

[1] Nicholas *et al.*, *Nat. Chem.*, **16**, 36–41 (2024).

[2] Michel *et al.*, *Chem. Mater.*, **20**, 4720–4728 (2008).

[3] Bennett *et al.*, *Phys. Rev. Lett.*, **104**, 115503 (2010).

[4] Raiteri *et al.*, *J. Phys. Chem. C*, **114**, 5997–6010 (2010).

[5] Drautz, *Phys. Rev. B*, **99**, 014104 (2019).