Predicting the atomic-scale structure of disordered materials with machine-learning potentials and experimental constraints

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Due to the lack of short-range order, the study of disordered materials has fallen somewhat beyond the scope of density functional theory (DFT), which is, on the other hand, routinely used to model crystals and small molecules. Since the recent introduction of machine learning potentials (MLPs) with quasi-DFT accuracy and low CPU cost, the atomic-level description of disordered and amorphous materials has taken both quantitative and qualitative leaps forward. Quantitatively, MLPs grant us access to time and length scales previously only accessible to empirical force fields, which are unable to reliably describe chemical reactions and diverse atomic environments. Qualitatively, the increase in accuracy and scale have unlocked simulations that were previously unfeasible, allowing us to rethink the way that we do atomistic modeling.

For the atomistic simulation of disorder materials, a crucial (and rather nontrivial) first step is the generation of realistic structural models, from which material properties can then be derived. A major challenge in this field has been how to reconcile simulation results with experiment. One way to do this is by generating many candidate structures according to some simulation protocol, like the so-called “melt-quench” procedure often used to generate structural models of amorphous solids. Unfortunately, structures generated this way tend to disagree with available experimental data. Another way is to try and mimic the experimental growth process explicitly, as has been done for amorphous carbon [1]. However, most of the time this kind of simulation is simply intractable, even with MLPs, due to inaccessible experimental time scales and otherwise extreme complexity (e.g., if chemical reactions are involved).

A third route is the generation of atomistic structures that are compatible with the experiment by design. This involves moving the atoms in such a way that the simulated and experimental target observables, e.g., an X-ray diffraction pattern, agree with each other. Reverse Monte Carlo is an example of such technique [2]. To ensure the generated structures remain physically sound, one can further enforce physical constraints by adding information about the potential energy of the system, i.e., to prevent the generation of high-energy structures, as in hybrid reverse Monte Carlo [3]. With the emergence of MLPs, these optimization schemes can now be performed with significantly higher fidelity than before, and for more chemically complex systems. At the same time, the availability of ML-based surrogate models to efficiently and accurately predict material observables amenable to direct comparison with experiment, like X-ray photoelectron spectroscopy (XPS) [4], opens the door to adding a wide variety of experimental constraints into the optimization.

In this talk, I will tell the story of how we put these ingredients together to overcome the existing challenges and predict the structure of oxygenated amorphous carbon (Fig. 1) [5], and how our group is currently refining and extending these techniques for higher versatility and computational efficiency.

References

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Figures

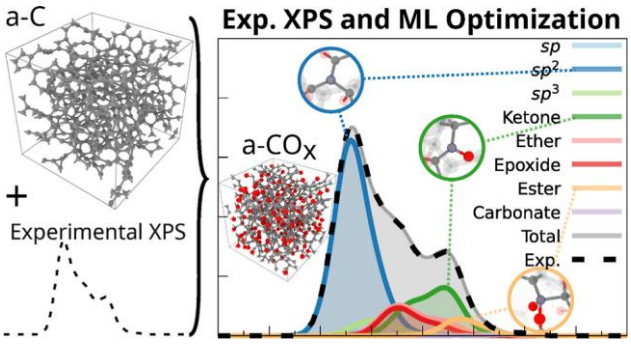

Figure 1: XPS deconvolution of an oxygenated amorphous carbon structure generated computationally to match experimental data, from Ref. [5].