Towards a general approach to the simulation of structural transformations and chemical reactions in condensed phases

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Computer simulations of transformation processes like phase transition and chemical reactions are an important complement to experiments, as they allow to understand mechanisms and to reconstruct free energy profiles and kinetic rates. We recently developed distance definitions able to quantify the similarity between atomic structures of solid materials as well as liquids and solutions. Next, we employed such distances to formulate new "reaction coordinates" that, in combination with enhanced sampling methods, allow the efficient reconstruction of free energy landscapes and barriers from *ab initio* and classical molecular dynamics simulations.

In materials science, our approach proved capable of resolving a number of crystalline polymorphs, amorphous forms and liquids over a range of ionic, covalent, metallic and molecular materials. Furthermore, we could recently reconstruct several challenging transitions of water (described with the TIP4P/2005 model) both at low and high pressure, obtaining in particular the crystallization of liquid and amorphous water without supercooling – in fact, even above the melting temperature, i.e., in unfavorable thermodynamic conditions. In the future, we hope to bridge the gap between the rich information provided by crystal structure prediction tools and the poor understanding of viable kinetic routes for the synthesis of new materials. In chemistry, our techniques allow to study chemical reactions with *ab initio* molecular dynamics passing in a seamless way from gas-phase to (explicit) solvent environments. Recent applications include the discovery of a possible prebiotic pathway to the hydrothermal synthesis of purine and pyrimidine ribonucleotides, and the rationalization of hydrothermal decomposition pathways of glycine and isovaline.

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