A novel numerical method for exploring challenging phase-transitions: From liquid-crystal to amorphous-amorphous transformations

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Abstract

Atomistic simulations, such as Molecular Dynamics, are powerful computational tools for investigating both ordered and disordered systems. However, in many situations, in particular those involving a kinetic barrier, one faces the *time scale problem*: the simulated time length is much too short to observe important phenomena (e.g. crystallisation from the liquid, relaxation in the vitreous state, etc). Thus, *enhanced sampling* methods (such as metadynamics) have been developed in the last decades and have proven efficient in many specific cases. However, a general and transferable method, that would reveal simultaneously the atomistic mechanism and the energetics of transformations has been lacking.
Water is a challenging system in this context because of its rich poly(a)morphism. Here, we show that using a novel set of coordinates, capturing changes in the topology of the interatomic network, we are able to systematically track transitions among liquid, amorphous and crystalline forms throughout the whole phase diagram of water [1], including the nucleation of crystals above and below the melting point. Our general approach is not specific to water and we will address on-going works in silica and B2O3. Both systems show a rich polymorphism as well as possible liquid-liquid transitions [2, 3]. The methodology will be used to establish connections between the liquid, glass and crystalline phases.

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