

## Metadynamical approaches to atomistic simulation of glasses

We employ metadynamics techniques, coupled with *ab initio* molecular dynamics (MD) simulations, to model defect-free atomistic configurations of hydrogenated amorphous silicon (*a*-Si:H), containing thousands of atoms. Metadynamics is a powerful non-equilibrium MD method, which can accelerate the sampling of atomistic configurations from the multidimensional free-energy surfaces (FES), associated with the (rare) events on a large simulation time scale, which are difficult to achieve via conventional MD simulations in phase space. This is generally performed by adding an external time-dependent bias potential, which is a function of a set of collective variables,  $S$ , to the Hamiltonian of the system. The collective variable  $S$  is defined as a generic function of the system coordinates (e.g., bond distance, bond angle, etc.) that is capable of describing the mechanism of interest. The bias potential discourages the system from sampling previously visited values of  $S$  (i.e., encourages the system to explore new regions of the FES) and, thus, it facilitates the system to cross barriers faster than that would occur in conventional MD simulations.

In our project, we successfully completed the development of atomistic models of disordered solids using metadynamical simulations, with an emphasis on tetrahedral amorphous semiconductors. Metadynamics simulations offer a systematic way to incorporate characteristic structural properties in a model by using a set of collective variables. By using appropriate collective variables in metadynamic simulations, it was demonstrated that high-quality models of *a*-Si:H can be produced with a hydrogen concentration ranging from 7 at.% to 22 at.%, making it possible to study the physics of device-grade *a*-Si:H for photovoltaic modules. A unique advantage of the method is that the concentration of H atoms in simulated samples can be controlled quite accurately by means of a set of collective variables.