

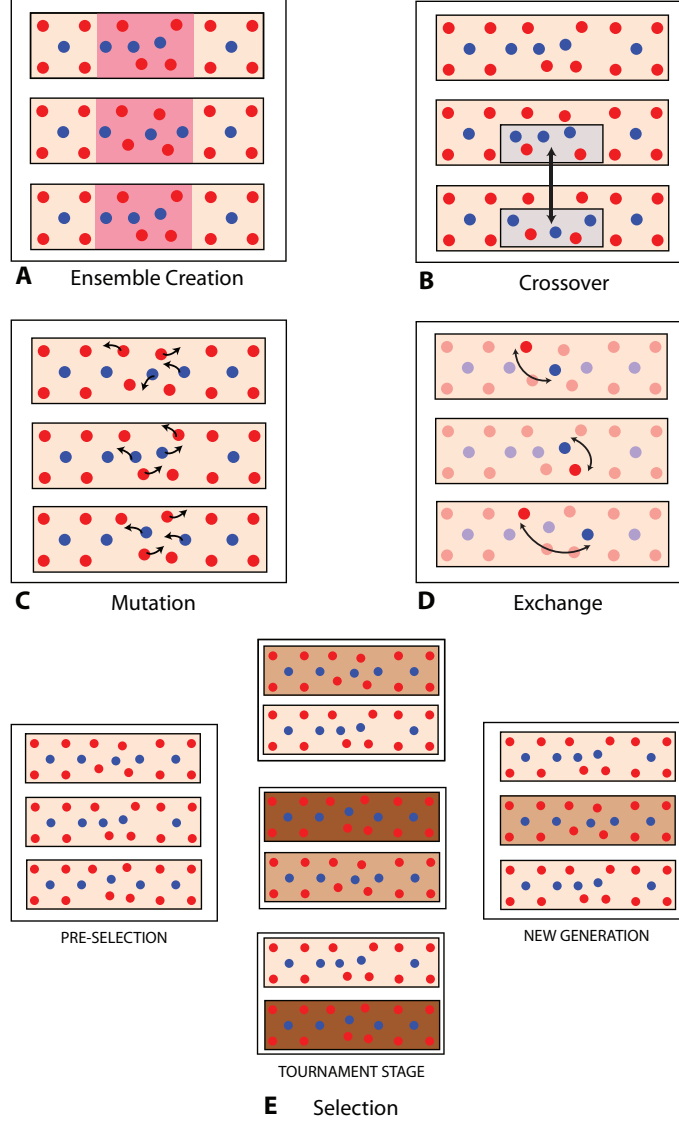


FIG. 1: **Schematic representation of the genetic algorithm.** (A) An ensemble of supercells containing the grain boundaries is created. The randomized regions in each supercell are shaded pink and are ‘enclosed’ on both sides by undisturbed, bulk crystal. (B) During crossover, groups of atoms are exchanged between randomly chosen pairs of configurations. The shaded boxes represent the cuboids described in the text. (C) Atoms are displaced by an amount inversely proportional to their order parameter during the mutation step. Although the diagram implies that only atoms in the disordered region are displaced, atoms in the bulk crystal region also move. Their displacements are much smaller however, owing to their much higher order parameter. (D) Two atoms to be exchanged in a configuration are chosen with probability proportional to their disorder. (E) The next generation of individuals is created using tournament selection. In the diagram, the tournament size is two and darker levels of shading indicate higher energy configurations. The ‘winner’ (lower energy configuration) of each tournament becomes a member of the next generation.

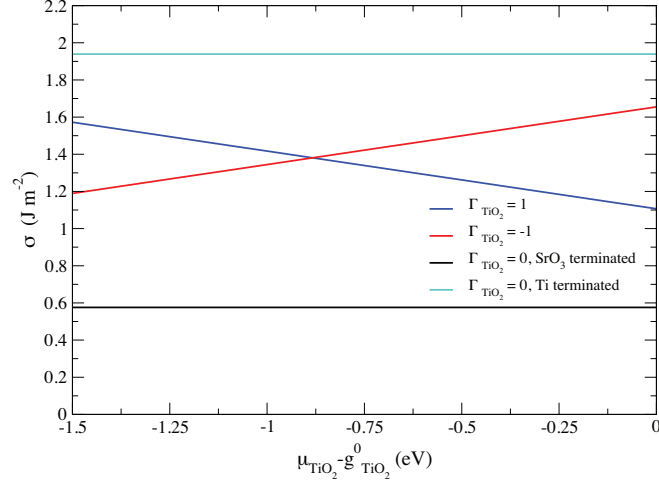


FIG. 2: Grain boundary energies as a function of μ_{TiO_2} for the $\Sigma 3(111)[\bar{1}10]$ interface. The area A_s of the supercell parallel the boundary plane has been set equal to 1 in specifying the boundary excesses Γ_i .

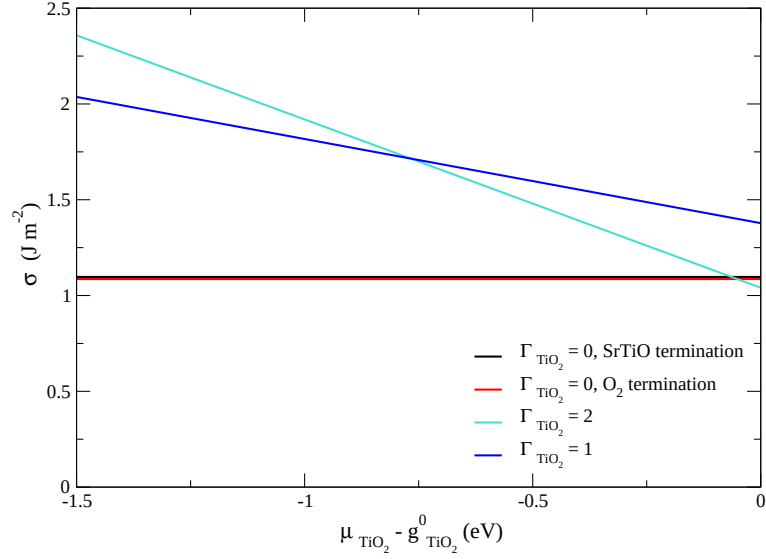


FIG. 3: Grain boundary free energies as a function of μ_{TiO_2} for the $\Sigma 3(112)[\bar{1}10]$ system.

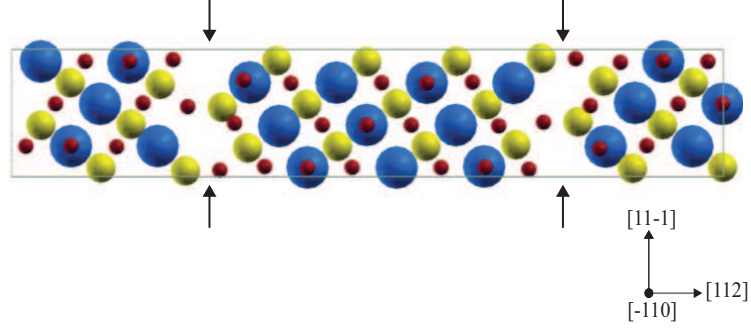


FIG. 4: The $\Gamma_{\text{TiO}_2} = 2 \Sigma 3(112)[\bar{1}10]$ grain boundary. The arrows indicate the position of the two equivalent grain boundaries in the supercell. Blue spheres indicate Sr atoms, red spheres indicate O atoms and yellow spheres indicate Ti atoms. The Ti and O atoms facing each other at the interface are in the same plane along the $[\bar{1}10]$ tilt axis.