Microscopic processes for hydrogen chemisorbed on graphene: permeation and recombination

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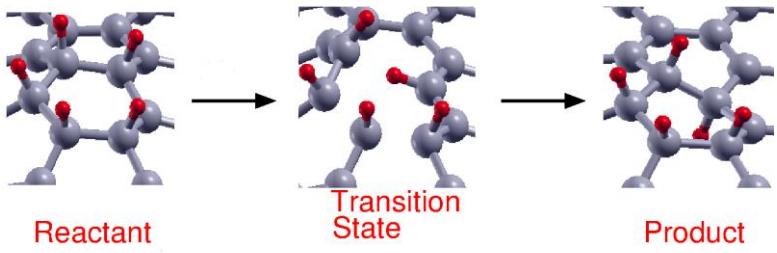
Hydrogenated graphene is of great interest in several different fields such as hydrogen technologies, astrochemistry, nuclear fusion, electronics and magnetism. Using large molecular prototypes of graphene, we have carried out density functional theory computations to study in detail two processes occurring in this system, namely, the permeation or flipping of chemisorbed hydrogen atoms through graphene and the recombination of these atoms (desorption leading to the formation of hydrogen molecules), which can be regarded as the subsequent step after permeation. Firstly, we will present a new mechanism for the flipping of chemisorbed hydrogen atoms[1] or protons[2] through a graphene layer (see figure), where we have found that the activation energies involved are of the order of recent experimental findings[3,4]. In addition, we will show preliminary results on reaction paths and rate coefficients for the recombination of hydrogen and deuterium, which exhibit large isotopic substitution effects (due to zero-point energy and tunneling) in qualitative agreement with thermal desorption measurements[5,6]. We believe that these studies will help to rationalize the experimental results as well as to provide some clues about properties of hydrogenated graphene.



Stationary point geometries of the reactant, transition and product states for a hydrogen atom flipping through a five-times hydrogenated carbon ring. The activation energy is about 1.5 eV.

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