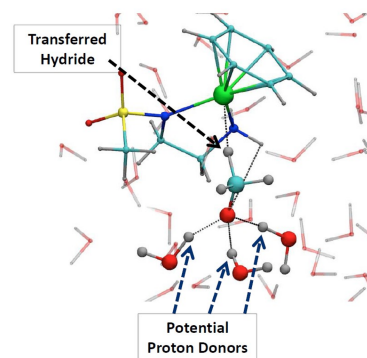


Modeling Chemical Reactions in an Aqueous Environment

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Understanding the mechanism and kinetics of chemical reactions in a complex fluctuating environment is a challenge. We demonstrate how *ab initio* molecular dynamics of chemical reactions in aqueous solution provides novel insight into the role of solvent molecules in the mechanism and kinetics of reactions. In our approach we combine a density functional theory (DFT) description of the electronic structure with advanced free-energy and path-sampling molecular dynamics techniques. This provides an accurate and realistic picture of the thermodynamics, mechanism and kinetics of the reaction pathways. For selected proto-type example reactions [1,2,3,4] we show how solvent water molecules play an active role in the reaction by participating via hydrogen bonding and mediating proton-transfer processes. This yields a picture of the free-energy profile and reaction mechanism that is fundamentally different from that predicted by gas-phase calculations without explicit solvent.



Solvent molecules play an active role in ruthenium-catalyzed conversion of formaldehyde to methanol.

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