Universität Basel

Research Project

Intermolecular Interactions and the Role of Dynamics for Chemical Reactions in Complex Systems

Third-party funded project

Project title Intermolecular Interactions and the Role of Dynamics for Chemical Reactions in Complex Systems
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The goal of this project is to develop, implement and applycomputational strategies to characterize, understand and predictproperties of complex systems at a molecular level. To this end,computational techniques including multipolar (MTP) force fields,adiabatic reactive molecular dynamics (ARMD) simulations, andmolecular mechanics with proton transfer (MMPT) are used and furtherdeveloped. Multipolar force fields will be extended to routinespectroscopic applications such as 1- and 2-dimensional infraredspectroscopies. This will be applied to spectroscopic probes,primarily nitriles (-CN), which are sensitive environmental probes forprotein interiors. Fluctuating MTPs will be used to study the 1d- and2d-IR spectroscopy of-CN-containing inhibitors in human aldosereductase (hALR2) and benzonitrile in Lysozyme. Multiplesurface-ARMD will be combined with the fewest switching surfacehopping (FSSH) methodology for investigating nonadiabatic effects inchemical dynamics. This will considerably extend the range ofapplicability for MS-ARMD. Initial applications concernphotodissociation and recombination of ICN in solution and O2formation in amorphous ice at low temperatures. In a next step, thestructural and solvent dynamics upon oxidation from Cu(I) to Cu(II) incopper-phenanthroline complexes will be investigated. Also, standardARMD simulations will be used to study multiple-ligand dynamics in theactive site of truncated Hemoglobin N which is responsible fordenitrification. Molecular mechanics with proton transfer will becombined with multi state-ARMD to investigate proton transfer in thecondensed phase on extended time scales. Computationally efficientevaluation of accurate energy functions is particularly important whenusing it for multidimensional spectroscopy which typically requiresextensive conformational sampling to converge the frequency frequencycorrelation function. The developments will allow atomistically refined simulations of the recently recorded 2d-IR spectrum of theexcess proton in liquid water. The present proposal involves tworesearch themes: reactive simulations in the condensed phase andcomputational vibrational spectroscopy which both require accurate representations of the intermolecular interactions.

Keywords Reactive Dynamics; QM/MM Simulations; Protein-Ligand Interactions; Computational Vibrational Spectroscopy; Multipolar Force Fields; Force Fields; Molecular Dynamics
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