

Welcome to the Doltsinis Group

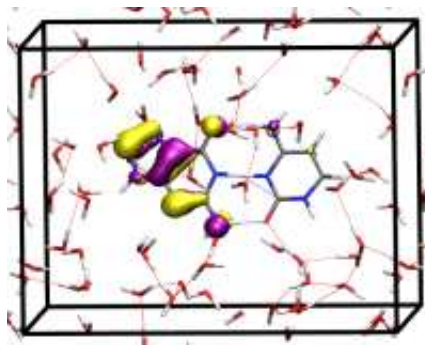
(Note that the Doltsinis Group has moved to Universität Münster)

Ab initio photochemistry of complex systems:

Photochemical reactions of large molecules, in particular in solution, can be studied using our recently developed nonadiabatic ab initio molecular dynamics method (na-AIMD) taking into account all nuclear degrees of freedom.

- **Applications:**

- The photoisomerisation of azobenzene derivatives plays an important role in materials science for the development of optical storage media, optical switches, and light-powered nanomachines.
- Knowledge of the photophysical properties of DNA bases and base pairs is the key to understanding the mechanisms leading to radiation induced genetic damage. Changes in the molecular structure following photoexcitation as well as the route for subsequent radiationless decay can provide important clues.



- **Method development:**

- Extension of na-AIMD to multiple electronic states using TDDFT. An ongoing collaboration with Prof. Kosov (Maryland) on the development of analytical gradients for TDDFT excited states as well as nonadiabatic coupling elements has existed for several years.

Molecular aggregation through hydrogen bonding:

Within the DFG funded Research Collaboration FOR 618 "Aggregation of small molecules" we study amino acids in aqueous solution using AIMD. Apart from conformational changes we investigate the neutral-zwitterionic transition as a function of the degree of solvation. Furthermore, we plan to use AIMD to study the co-crystallization in formamide-water mixtures. Our calculations are accompanied by the experimental efforts of Prof. Kleinermanns (Düsseldorf), Profs. Sander and Havenith (Bochum) and Prof. Boese (Essen).

Diffusion and speciation of dissolved oxides in H₂O at high pressures and temperatures:

This project is carried out in collaboration with Prof. Maresch (Institut für Geologie, Mineralogie und Geophysik, Bochum) within the Collaborative Research Centre (SFB) 526 "Rheology of the Earth - from the Upper Crust to the Subduction Zone". Speciation and diffusion of quartz and other minerals in water at high temperatures and pressures will be investigated theoretically by means of AIMD simulations as well as experimentally using Raman spectroscopy and Fabry-Perot interferometry.

Diffusion processes at solid-solid-liquid interfaces:

This project is carried out in collaboration with Prof. Renner (Institut für Geologie, Mineralogie und Geophysik, Bochum) within the Collaborative Research Centre (SFB) 526 "Rheology of the Earth - from the Upper Crust to the Subduction Zone". Diffusion processes and surface reactions potentially leading to groove formation at the quartz-water interface will be studied using AIMD and AFM measurements.

Novel constraint methods for free energy and rare events:

- **Applications:** Study of tautomerisation of DNA bases and base pairs in ground and excited state, in particular in aqueous solution using **coordination constraints**. Goal: Understanding of genetic damage caused by the formation of so-called rare tautomers.
 - **Method development:** Development and implementation of novel constraint methods within the framework of AIMD. The aim is to search for low energy reaction paths on complex free energy landscapes and to be able to reconstruct the latter at least partially. Our recent developments along these lines include the **Dynamic Distance** method and **Targeted AIMD**. Going beyond a purely ab initio implementation we plan to extend the applicability of these methods to large biochemical systems by using a hybrid quantum-classical QM/MM approach. This work has been carried out in collaboration with Dr. Markwick (Grenoble), Prof. Marx (Bochum) and Prof. Schlitter (Bochum).
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Developments within the CPMD package:

- nonadiabatic dynamics (ab initio surface hopping, QM/MM extension)
 - new geometric constraints (Targeted MD, Dynamic Distance Constraint)
 - time-dependent density functional theory (TDDFT excitation energies, gradients, and nonadiabatic couplings)
 - new density functionals (HCTH family)
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Publications

Group Members