Non-Adiabatic Charge Transfer Simulation of Photoactivation in E.Coli DNA Photolyase

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DNA Photorepair

Photolyases bind to UV-damaged DNA and repair CPD lesions via electron transfer from the FAD cofactor to the dimerized pyrimidines. During photoactivation an electron is transferred from the protein surface to the partially oxidized flavin cofactor.

- dynamic and heterogeneous environment
- transfer over long distances and long timescales
- Rate of transfer ≈ environmental reorganization energy

Multi-Scale Simulation Scheme

Coarse Graining of quantum system

QM/MM Embedding

- Polarisation of QM system by environment $H_{\text{env}}$ is evaluated with QM/MM-environment represented by point charges
- Influence of changing charge distribution on classical environment: Mapping fraction of fragment charge onto atomic charges
- Charge resides mainly on Trps
- On average charge transfer occurs within 50 ps
- Small partial occupation of Trp 316
- Drop in ESP leads to drop in IP
- Transfer occurs within 25 ps resp 10 ps for the two scaling factors

Electronic Polarisation of environment

- Nonpolarizable forcefields can lead to overestimation of environmental reorganisation energy.
- Scaling of MM charges by suitable constant

Summary and Outlook

- Trp 316 might be involved in the transfer process; originally not considered in transfer path
- ESP scaling is important in order to reproduce experimental values
- Charge transfer dynamics can be reproduced very well without prior assumption of the involved amino acids
- New charge distribution on carriers
- Starting structure: MD snapshot
- MM step: Perform classical MD step
- New charge distribution on carriers
- Calculation of electronic structure of cc

References:
1. Woiczikowski et al. JPCB (2011) 115, 9846
2. Kubar and Elstner JPCB (2010) 114, 11221