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Molecular Dynamics and Computational Spectroscopy for Systems in Soft Condensed Phases: Biological Macromolecules, Molecular Aggregates & Ions in Solution

Molecular dynamics is a wide-spread simulation technique, which (i) I have employed to elucidate photchemical and -physical processes based on potentials from electronic structure calculations1,2 and (ii) can be used to obtain structural insights into biological macromolecules like proteins and DNA using classical force fields. Complementary to this, computational molecular spectroscopy allows to establish structure-property relationships. For example, my calculations aided in assigning chemical shifts and hyperfine couplings to atoms of chromophores inside proteins.3,4 In my research, I have also combined both in the framework of quantum mechanics/molecular mechanics (QM/MM) simulations to understand the molecular origin of the difference in optical absorption between the photoproduct (Pg) and dark state (Pr) of the photoreceptor protein Slr1393.5 Furthermore, my QM/MM-optimized structures of Pr, Pg, and a photointermediate together with recent experimental advancements have provided further atomistic details concerning structural rearrangements in the chromophore-binding pocket along the photocycle.6
Here in Augsburg, my PhD student and I are working on unravelling the mechanisms leading to thermally activated delayed fluorescence in donor-acceptor cyanoarenes7 by employing QM and QM/MM to describe the properties of these molecules in molecular aggregates. A project that is funded in the framework of the HYP*MOL TRR. I also perform simulations of biological macromolecules in solution. For this purpose, I am working on the parameterization of metal ions, e.g. Mn2+, which is a prerequisite for classical MD simulations. This will pave the way towards understanding the influence of salt concentration on the structure of biological macromolecules.

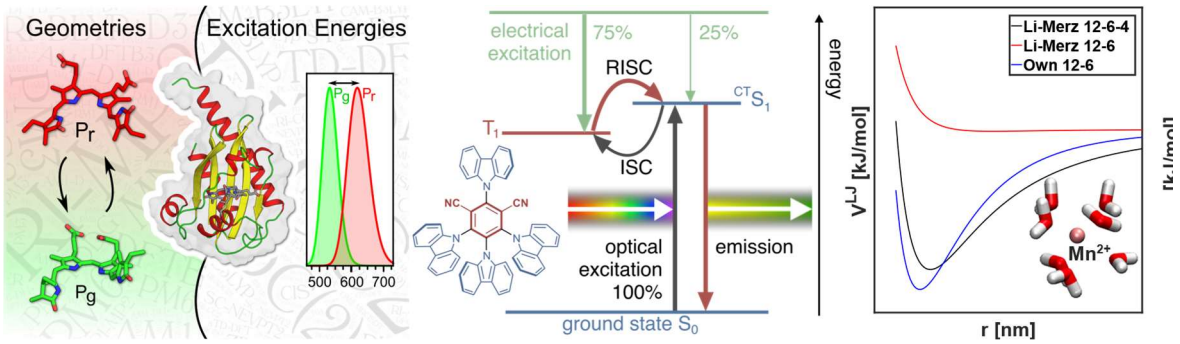

Fig. 1 – left: Scheme showing molecular structures of the phycocyanobilin chromophore inside Slr1393 from QM/MM simulations and corresponding absorption spectra; middle: Illustration of relevant processes leading to thermally activated delayed fluorescence in 4CzIPN; right: Lennard-Jones potential of Mn^{2+} and its coordination in water shown as inset.

References

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