Multiscale Modeling of Structural and Optical Properties of Complex Supramolecular Aggregates

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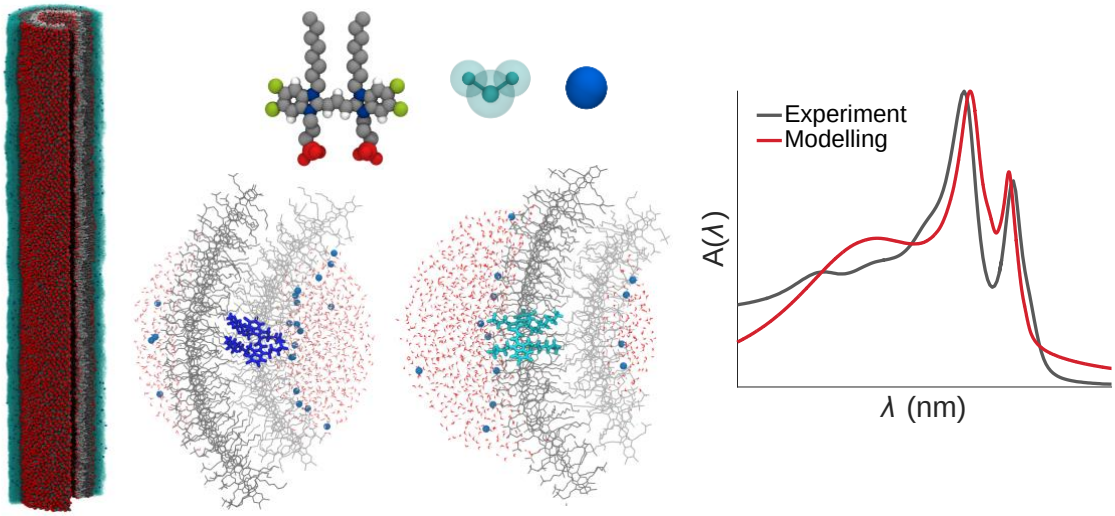
High interest in molecular tubular C8S3 J-aggregates[1,2] arises from their structural resemblance to the light-harvesting complex of green sulfur bacteria – the most efficient photosynthetic organisms – as well as their potential as energy transport wires. These functionalities are governed by the excitonic spectrum, which in turn sensitively depends on molecular packing. As the structure cannot be resolved experimentally, theoretical modeling is important.

Here, we report on a new level of first-principles modeling of the optical properties of these synthetic amphiphile aggregates[3]. We use the results of large-scale molecular dynamics (MD) simulations for the structure, which is translated into an exciton Hamiltonian. MD snapshots provide information on the effect of disorder and on the interaction of the dye molecules with the solvent. Polarizable force field calculations shed light on the different electronic environments felt by the inner and outer wall molecules. By modeling the absorption spectrum and comparing to the experimental one, we verify the validity of the obtained structure.



[1] D Eisele, C Cone, E Bloemsma, *et al., Nat. Chem.* 4, **2012**

[2] B Kriete, AS Bondarenko, VR Jumde, *et al., J. Phys. Chem. Lett.* 8, **2017**

[3] AS Bondarenko, I Patmanidis, R Alessandri, Paulo C.T. Souza, TLC Jansen, AdH Vries, SJ Marrink, and J Knoester, *Chem. Sci.,* 11, **2020**