Reaction-Diffusion Simulations of Photoredox Processes in Solution

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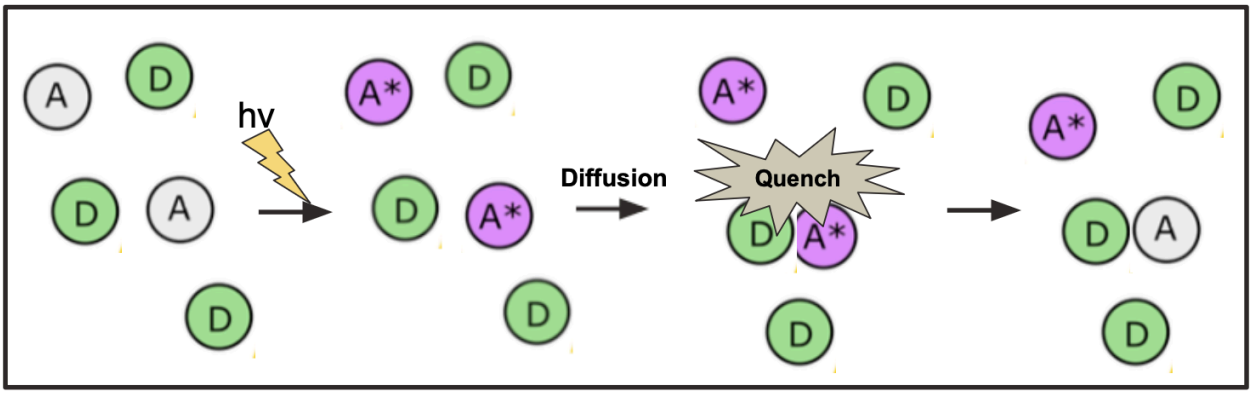
A computational method to simulate bimolecular quenching reactions in solution using coarse-grained reaction-diffusion dynamics is presented and applied to quenching of molecular photosensitizers in solutions.[1,2,3,4] The simulations are implemented to describe photoinduced reactions involving explicit excited states of light-harvesting species, that can be populated by a pulsed excitation, together with intrinsic deactivation as well as collision quenching from separate quencher species (Scheme 1). The presented simulation methodology is applied to quenching of light-harvesting Fe(III) complexes in electron donating solvents over a wide range of quencher concentrations as a prototype system of experimental interest for the reaction-diffusion dynamics over a wide range of concentrations.[3] The results show clear signatures for the transition from classical diffusion-limited Stern-Volmer dynamics to close-contact quencher-photosensitizer interactions at high quencher concentrations, and the simulations are used to assess physically realistic photosensitizer-quenching collision interaction parameters for photoinduced dynamics beyond the classical Stern-Volmer model.

Scheme 1: Collision quenching of donor and excited acceptor



Acknowledgements:

The Swedish Research Council (VR, 2021-05313), the Knut and Alice Wallenberg (KAW, 2016.0059) foundation, the Swedish e-Science initiative eSSENCE, and NanoLund are acknowledged for support. Computing resources were generously provided by the LUNARC and NSC high performance computing centers via SNIC/NAISS allocations. We thank Prof. Arkady Yartsev for valuable discussions.

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