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Tensor network methods for the simulation of open quantum dynamics in multichromophore systems: Application to singlet fission in novel pentacene dimers[1]

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Singlet fission (SF) is an ultrafast process in which a singlet exciton spontaneously converts into a pair of entangled triplet excitons on neighbouring organic molecules. As a mechanism of multiple exciton generation, it has been suggested as a way to increase the efficiency of organic photovoltaic devices, and its underlying photophysics across a wide range of molecules and materials has attracted significant theoretical attention. Recently, a number of studies using ultrafast nonlinear optics have underscored the importance of intramolecular vibrational dynamics in efficient SF systems, prompting a need for methods capable of simulating open quantum dynamics in the presence of highly structured and strongly coupled environments. Here, a combination of ab initio electronic structure techniques and a new tensor-network methodology for simulating open vibronic dynamics is presented and applied to a recently synthesised dimer of pentacene (DP-Mes). We show that ultrafast (300 fs) SF in this system is driven entirely by symmetry breaking vibrations, and our many-body approach enables the real-time identification and tracking of the "functional' vibrational dynamics and the role of the "bath"-like parts of the environment. Deeper analysis of the emerging wave functions points to interesting links between the time at which parts of the environment become relevant to the SF process and the optimal topology of the tensor networks, highlighting the additional insight provided by moving the problem into the natural language of correlated quantum states and how this could lead to simulations of much larger multichromophore systems

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