Photoreactions With Quantum Treatment of Coupled Electronic, Photonic, and Nuclear Degrees of Freedom

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This project is part of an effort to create distributable software that can be used to explore light-driven chemical reactions. More specifically, it is an attempt to model photoreactions under a regime that includes quantum treatment of coupled electronic, photonic, and nuclear degrees of freedom. The goal is to do this in such a way that freely allows for energy to be exchanged back and forth between these degrees of freedom. This is so that these interactions can be monitored and observed in a quantifiable manner, and thereby predicted for real systems. This computational tool is being developed with a broad range of applications in mind. The first milestone will be a tool to screen photodissociation of diatomic molecules in a cavity resonator, providing an estimate of the yield of the reactions. A comparison of the findings to the available experimental data will serve to validate the method. Upon validation, this tool will be used to model and predict ultrafast photoreactions of molecules for which there is not yet experimental data. This is in an effort to sustain a feedback loop between theory and experiment in the field of photochemistry. Another exciting and natural direction to develop this tool is the modeling of exciton-polaritons. This capability, along with the inevitable numerous incremental steps toward achieving it, has the potential to greatly elucidate important factors in the creation, sustainability, and termination of exciton-polaritons.

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