Mixed Quantum-Classical Methods for Molecules in Cavities

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Describing chemical processes that are strongly correlated with quantum light requires an accurate, flexible, and computationally efficient treatment of light-matter interactions. Thus, in order to develop ab-initio theoretical descriptions of cavity modified chemical systems, extensions to the traditional theoretical tool-kits for quantum optics and quantum chemistry are required. Here, we investigate the extension of mixed quantum-classical trajectory methods as well as the concept of time-dependent potential energy surfaces, both traditionally introduced for electron-nuclear problems, to the photonic degrees of freedom. Within our work we find that classical Wigner dynamics for photons can be used to describe quantum effects such as correlation functions, bound photon states and cavity induced suppression by properly accounting for the quantum statistics of the vacuum field while using classical/semi-classical trajectories to describe the time-evolution. Investigating the time-dependent potential energy surfaces for photons we find significant differences with the potential used in conventional approaches. Furthermore, we analyze the exact time-dependent potential energy surface driving the proton motion of a cavity-induced chemical suppression.

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