Physical Chemistry Seminar

First-Principles Quantum Electrodynamics Methods for Polariton Chemistry

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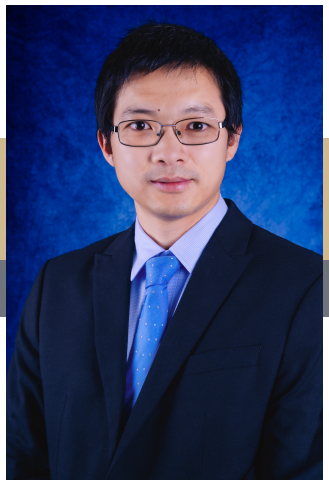

The intersection of quantum electrodynamics (QED) and molecular processes has yielded remarkable advancements in altering molecular properties and reactivity by exploiting light-matter couplings. As polaritonic states—hybrid electron-photon states—gain prominence in chemical applications, the need for understanding the underlying mechanisms of polaritons-mediated processes has stimulated the development of new ab initio methodologies. These advancements have led to the extension of many traditional electronic structure methods to molecular quantum electrodynamics framework, including Hartree-Fock, density functional theory, and coupled-cluster theories. In this talk, I will introduce a variational transformation-based molecular quantum electrodynamics mean-field method (VT-QEDHF) designed to handle light-matter interactions across all coupling strengths. This method naturally bridges the gap between approaches like QED Hartree-Fock in the weak coupling regime and strong coupling QEDHF methods, demonstrating superior versatility and reliability. Additionally, I will present the quantum Monte Carlo (QMC) method for obtaining numerically exact solutions for polaritonic ground states during the dissociation of the H_2 molecule. Our study reveals significant insights into electron-nuclear-photon interactions, such as shifts in the cavity Born-Oppenheimer surface minimum, wave function localization, and mode occupation, with results compared directly against state-of-the-art polaritonic coupled-cluster methods. Together, these advancements mark significant progress in ab initio modeling of polariton chemistry, offering new pathways for understanding and manipulating light-matter interactions at the quantum level.

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