

2024 年度年次報告書

量子・古典の異分野融合による共創型フロンティアの開拓

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Mixed Quantum-Classical Multiscale Theoretical Study of Molecular Polariton Physics and
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研究成果の概要

Molecular exciton polaritons are a captivating class of hybrid light-matter states that emerge from the strong coupling between molecular electronic excitations and optical cavity modes. By modulating the strength of light-matter coupling, it becomes possible to reshape the energy landscape and influence the dynamic behavior of molecules, opening new avenues for applications in energy conversion, molecular electronics, and catalysis. Experimental studies have revealed their profound impact on diverse processes, including the modulation of reaction kinetics, the enhancement or suppression of energy transfer pathways, and even the inversion of excited-state populations.

From a theoretical standpoint, strong light-matter interactions give rise to a range of collective and quantum effects that are absent in conventional molecular systems. Despite their intriguing potential, accurately modeling these systems poses significant computational challenges due to the vast number of molecular degrees of freedom involved and the intricate nature of their interactions with the cavity mode.

In the fiscal year 2024, I developed a semiclassical truncated-Wigner-approximation (TWA) theory that focuses on the dynamics of molecular exciton polaritons [J. Chem. Theory Comput. 21, 1509-1520 (2025)]. This work constitutes a noteworthy advancement of the TWA theory of molecular vibration-polariton dynamics [J. Chem. Theory Comput. 20, 3019-3027 (2024)], necessitating the inclusion of quantum coherence between the electronic ground and excited states of the molecules in a semiclassical manner. To validate the TWA in this context, we first apply the theory to a simplified model where molecules are treated as two-level systems without vibronic coupling. By comparing semiclassical results to full quantum dynamic simulations, we demonstrate that the TWA provides accurate predictions for large molecular systems. This accuracy arises from the enhanced mean-field behavior and diminished influence of quantum correlations and nonlinearity in such extended systems of many emitters strongly coupled to a single cavity mode. Lastly, we extend the TWA framework by incorporating nuclear degrees of freedom and vibronic coupling to explore the decay of quantum coherence between electronic excitations in different molecules. Notably, we find that strong light-matter coupling mitigates the decay of quantum coherence, consistent with the dynamic polaron decoupling effect. These results underscore the robustness and versatility of the TWA in capturing key dynamics in strongly coupled exciton-polariton systems.

【代表的な原著論文情報】

- 1) Phuc, N. T. Semiclassical Truncated-Wigner-Approximation Theory of Molecular Exciton-Polariton Dynamics in Optical Cavities. *J. Chem. Theory. Comput.*, **21**, 1509-1520 (2025).