Multi-Scale Molecular Dynamics Simulations of Molecular Ensembles Strongly Coupled to Low-Quality Dispersive Optical Cavities

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Coupling molecules to the confined light modes of an optical cavity is showing great promise for manipulating chemistry and several examples of cavity-modified chemistry have been reported recently.[1-3] However, to fully exploit this principle and use cavities as a new tool for chemistry, a much better understanding of the effects of strong light-matter coupling on molecular dynamics is required. While quantum chemistry calculations provide atomistic insight into the reactivity of uncoupled molecules, the possibilities to also explore strongly coupled systems are still rather limited, because of the challenges associated with an accurate description of the cavity in these calculations. Despite recent progress in introducing strong coupling effects in quantum chemistry, applications are mostly restricted to single or simplified molecules in ideal lossless cavities that furthermore lack dispersion. In contrast, experiments are most often done with very many molecules in complex solvent environments inside low-quality Fabry-Pérot cavities. To close the gap between experiment and computation, we have extended the established Jaynes-Tavis-Cummings model into a multi-scale molecular dynamics framework, with which we can simulate ensembles of thousands of molecules and their environment,[4,5,6] strongly coupled to realistic cavities and track the dynamics of polaritons as a function of energy and *k*-vector. In the talk, we will briefly discuss this framework and illustrate the methodology with recent applications on polaritonic chemistry.

References

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