Accurate and Efficient Non-adiabatic Quantum Dynamics using Master Equations

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The exact treatment of real time nonadiabatic quantum dynamics in condensed phase chemical systems remains a significant challenge that spurs the ongoing development of approximate methods that are accurate, efficient, and can treat large systems with a wide range of different forms of interactions. Quantum-classical trajectory based methods provide some of the most appealing solutions to this problem that offer a hierarchy of approaches with different balances between accuracy and computational cost. However, since moving up this hierarchy typically requires orders of magnitude more computational effort, only the lowest tiers are likely to be practical, both now and in the foreseeable future, for nonadiabatic problems containing large quantum subsystems. In this talk I will discuss our recent research showing both how and why quantum-classical approaches can be made both more accurate and efficient by combining them with the formally exact quantum master equation framework. This combination of quantum-classical theory and master equation techniques makes it possible to obtain the accuracy of much more computationally expensive approaches at a cost an order of magnitude faster than even the most efficient trajectory based approaches, providing the ability to treat the quantum dynamics of atomistic condensed phase systems for long times.

References

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