Importance of Quantum Bath Sampling on Heat Transport in a Model Single-Molecule Junction

Pablo Carpio-Martínez and Gabriel Hanna

Department of Chemistry, University of Alberta, AB, Email: carpioma@ualberta.ca

Beyond the prototypical nonequilibrium spin-boson model (see Figure) of a single-molecule junction, the use of quantum bath sampling for calculating heat transport properties has been very limited due to its high computational cost. When simulating heat transport in realistic systems, it is therefore common to sample the initial bath conditions from classical probability distributions. In the present work, we study the effects of both quantum and classical sampling of the initial bath conditions on the steady-state heat current in the nonequilibrium spin-boson model. Over a wide range of parameter regimes, our findings show that the steady-state heat currents resulting from classical bath sampling are predominantly larger than those from quantum bath sampling. In both cases, the steady-state heat currents exhibit the expected turnovers as a function of the bath reorganization energy, albeit with different temperature dependencies of the turnover maxima. In general, as the bath temperatures are increased, the results of the classical and quantum bath sampling decrease but remain non-negligible at high temperatures. These differences are attributed to the more pronounced dependence of the classical bath distribution on temperature compared to the quantum one. Finally, quantum bath sampling is shown to be necessary for satisfying the steady-state fluctuation theorem – a microscopic generalization of the second law of thermodynamics. Overall, our results underscore the importance of quantum bath sampling in dynamical simulations of quantum heat transport.