Simulating 2D Vibrational and Electronic Spectroscopies: Hierarchical Equation of Motion (HEOM) Approach

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Multi-dimensional spectral profiles contain significant information on various dynamic processes, such as inter- and intramolecular couplings, and microscopic exiciton and electron transfers under the peculiarities of dissipative dynamics. The HEOM approach has a capability to simulate such dynamics, where the non-Markovian and non-perturbative system-environment interaction play a significant roles.[https://aip.scitation.org/doi/10.1063/5.0011599] Here, the theoretical basis of 2D spectroscopies on the basis of a Brownian and spin-Boson models with use of the HEOM are explained. If time allows, a possibility to measure free-energy by spectroscopic means that overcomes the difficulty of the Jarzynski approach [https://journals.jps.jp/doi/full/10.7566/JPSJ.90.033001] will be discussed.