View Abstract

CONTROL ID: 3318387
TITLE: Nonadiabatic Electron Nuclear Dynamics in TDDFT with Variational Quantum Nuclei
Abstract Body: Electron-nuclear coupling in non-equilibrium dynamics plays a fundamental role in condensed matter physics, defining behavior from phase transitions to chemical reactions. Recently the dynamics of these processes have been able to be experimentally resolved in the time-domain, requiring a theoretical framework beyond the Born-Oppenheimer approximation to describe [1]. While approaches such as the multi-configuration time-dependent Hartree method have been successful at describing excited state dynamics of relatively small systems, the cost to precompute potential energy surfaces and non-adiabatic couplings becomes a significant bottleneck for larger systems. In this talk we develop a trajectory based variational ansatz which treats the electronic system at the level of time-dependent density functional theory, while simultaneously incorporating a nonadiabatic quantum mechanical description of the nuclei. This method is compared to multi-trajectory Ehrenfest dynamics in the resolution of nuclear effects within the optical spectrum of small molecules.

[1] Kiyoshi Ueda et al 2019 J. Phys. B: At. Mol. Opt. Phys. 52 171001
Funding Acknowledgement: We acknowledge funding from the ERC through the QSpec-NewMat Project
PRESENTATION TYPE: Oral
UNIT: 05.0 CHEMICAL PHYSICS (DCP)
SORTING CATEGORY: 05.01.01 Electronic-vibrational coupling in light harvesting (DCP, DCOMP, DCMP, DPOLY, DAMOP) [same as 16.01.22, 01.01.44, 06.01.09]
Category Type: Computational
AUTHORS (FIRST NAME, LAST NAME): Kevin Lively[1], Guillermo Albareda[1], Aaron Kelly[2], Shunsuke Sato[3], Angel Rubio[1]
INSTITUTIONS (ALL): 1. Max Planck Inst Structure & Dynamics of Matter
2. Chemistry, Dalhousie University
3. Physics, University of Tsukuba
Teams: (none)