VISTA Seminar

Seminar 15

March 31, 2021

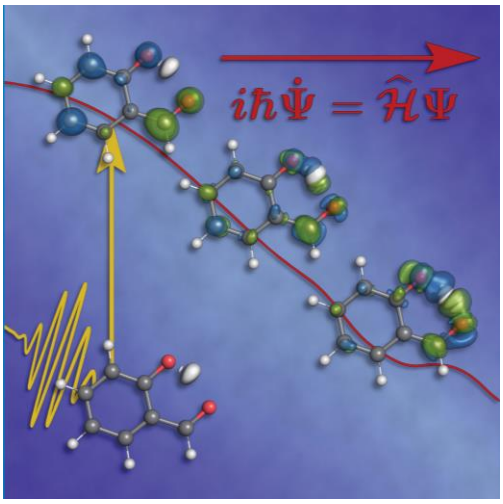
9:30 – 11:00 am EDT / 2:30 – 4:00 pm GMT / 3:30 pm – 5:00 pm Paris

TOC:

Ab Initio Nuclear-Electronic Orbital Ehrenfest Dynamics

Xiaosong Li

University of Washington | 206.685.1804 | http://faculty.washington.edu/xsli

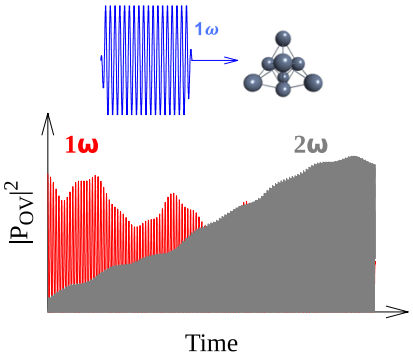


The real-time nuclear-electronic orbital (RT-NEO) approach, developed in collaboration with the Hammes-Schiffer group, provides an elegant framework for treating electrons and selected nuclei, typically protons, quantum mechanically in nonequilibrium dynamical processes. The dynamical interactions between the other nuclei and the electron-proton subsystem are described with the mixed quantum-classical NEO-Ehrenfest dynamics method. The NEO-Ehrenfest approach propagates the electrons and quantum protons in a time-dependent variational framework, while the remaining nuclei move classically on the corresponding average electron-proton vibronic surface. Our study shows that the NEO-Ehrenfest approach with a semiclassical traveling proton basis method yields accurate predictions of molecular vibrational frequencies. We also conduct NEO-Ehrenfest dynamics simulations to study an excited state intramolecular proton transfer process. These simulations reveal that nuclear quantum effects influence the predictions of proton transfer reaction rates and kinetic isotope effects due to the intrinsic delocalized nature of the quantum nuclear wave function. This work illustrates the importance of nuclear quantum effects in coupled nuclear-electronic dynamical processes and shows that the NEO-Ehrenfest approach can be a powerful tool to provide insights and predictions for these processes.

Ultrafast Nonlinear Plasmon Decay Processes in Silver Nanoclusters

Gowri Kuda-Singappulige

Department of Chemistry, Kansas State University, USA Email: gowir@ksu.edu



Uncovering the fundamental mechanisms behind plasmon mediated processes is the key to improve the efficiency of plasmonic materials. Light-induced processes in silver nanoparticles are of great interest, thanks to their strong absorption properties in the UV/Vis region. Due to the complexity and the ultrafast nature, nonradiative plasmon decay occurring at femtosecond time scale is challenging to resolve experimentally and theoretically. Real-time time-dependent density functional theory (RT-TDDFT) is an ideal computational tool to track electronic motion subjected to a perturbation. Recent studies[1-2] on silver nanoclusters suggest plasmon dephasing into hot carriers. Yan et al.[3] showed that the hot carriers generated by plasmon decay lead to nonlinear effects in silver atomic chains. Our work[4] focuses on the electron dynamics of a tetrahedral silver cluster, Ag_8 (T_d), upon excitation of plasmon-like states with a resonant continuous-wave electric field. We monitor the variation of off-diagonal density matrix elements corresponding to single-particle transitions in the Kohn-Sham orbital basis during the real-time propagation. Time variation, frequency analysis, and the assessment of symmetry-adopted selection rules for one- and two-photon allowed transitions demonstrate the rapid decay of the linear optical response and the emergence of non-linear optical properties in silver clusters as a result of plasmon decay. In addition, this work provides evidence that plasmonic character of materials play a significant role in enhancing nonlinear properties.

References:

1. Ma, J.; Wang, Z.; Wang, L.-W. Interplay Between Plasmon and Single-Particle Excitations in a Metal Nanocluster. *Nat. Commun.* **2015**, *6*, 10107/1-12.
2. Rossi, T. P.; Erhart, P.; Kuisma, M. Hot-Carrier Generation in Plasmonic Nanoparticles: The Importance of Atomic Structure. *ACS Nano* **2020**, *14*, 9963-9971.
3. Yan, L.; Guan, M.; Meng, S. Plasmon-Induced Nonlinear Response of Silver Atomic Chains. *Nanoscale* **2018**, *10*, 8600-8605.
4. Kuda-Singappulige, G. U.; Lingerfelt, D. B.; Li, X.; Aikens, C. M. Ultrafast Nonlinear Plasmon Decay Processes in Silver Nanoclusters. *J. Phys. Chem. C* **2020**, *124*, 20477-20487.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 15
Time: Mar 31, 2021 09:30 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/92312072622?pwd=K0RlRnMzOThNYjdrblFIVDk4Q2tEQT09

Meeting ID: 923 1207 2622
Passcode: 302753
One tap mobile
+16465588656,,92312072622#,,,,*302753# US (New York)
+13017158592,,92312072622#,,,,*302753# US (Washington DC)

Dial by your location
- +1 646 558 8656 US (New York)
- +1 301 715 8592 US (Washington DC)
- +1 312 626 6799 US (Chicago)
- +1 253 215 8782 US (Tacoma)
- +1 346 248 7799 US (Houston)
- +1 669 900 9128 US (San Jose)

Meeting ID: 923 1207 2622
Passcode: 302753
Find your local number: https://buffalo.zoom.us/u/adNOcVVoCK

Join by SIP
92312072622@zoomcrc.com

Join by H.323
162.255.37.11 (US West)
162.255.36.11 (US East)
115.114.131.7 (India Mumbai)
115.114.115.7 (India Hyderabad)
213.19.144.110 (Amsterdam Netherlands)
213.244.140.110 (Germany)
103.122.166.55 (Australia Sydney)
103.122.167.55 (Australia Melbourne)
149.137.40.110 (Singapore)
64.211.144.160 (Brazil)
69.174.57.160 (Canada Toronto)
65.39.152.160 (Canada Vancouver)
207.226.132.110 (Japan Tokyo)
149.137.24.110 (Japan Osaka)
Meeting ID: 923 1207 2622
Passcode: 302753