

VISTA Seminar

Seminar 1

9:30 – 11:00 am EDT / 1:30 – 3:00 pm GMT

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Branching Correction for Mixed Quantum-Classical Dynamics

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Abstract In chemistry, physics, biology, and material sciences, many important processes are relevant to nonadiabatic dynamics. Surface hopping and mean field have become the most widely used mixed quantum-classical methods for studying general charge and exciton dynamics. The reliability of surface hopping simulations relies on robust crossing and decoherence corrections.¹⁻⁴ Recently, we introduced a branching correction to surface hopping, which resets the wavefunction coefficients based on the judgement of wave packet reflection.⁵ Our method ensures the self-consistency of using the traditional time-dependent Schrödinger equation to describe the mixed quantum-classical dynamics and captures the majority of the decoherence effect as demonstrated in the three standard Tully models (see Figure 1) and over two hundred additional scattering models, surpassing other investigated decoherence correction algorithms. Inspired by the branching correction, we proposed better energy-based decoherence time formulas.⁶ We further applied a similar branching correction to mean field and achieved about 10-fold improvement of accuracy compared with the widely used Ehrenfest mean field method.⁷ In fact, the branching corrected mean field and surface hopping methods provide highly reliable and almost identical results using the exact quantum solutions as references in the investigated systems, implying that branching correction is essential in mixed quantum-classical dynamics and shows good potential for application in general nonadiabatic dynamics simulations.

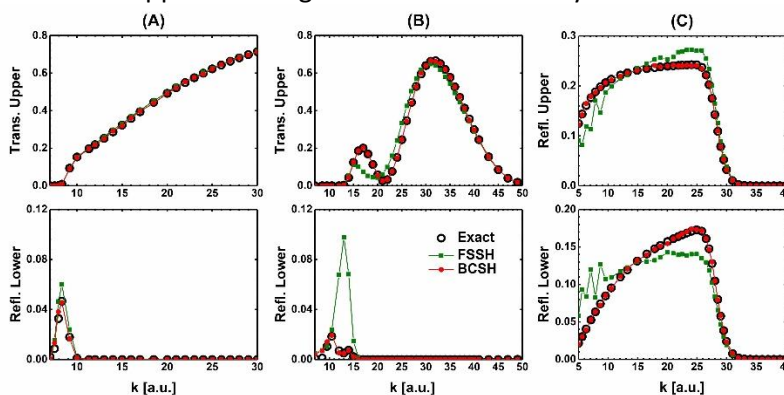
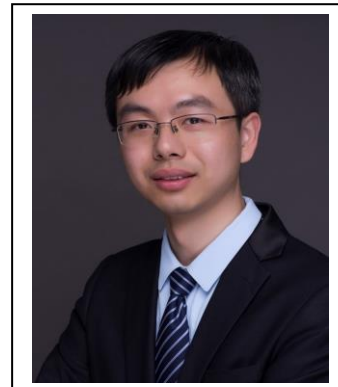


Figure 1. Performance of the fewest switches surface hopping (FSSH) and the branching corrected surface hopping (BCSH) in the three standard Tully models.

References

- (1) Wang, L.*; Akimov, A.; Prezhdo, O. V.* *J. Phys. Chem. Lett.* **7**, 2100 (2016).
- (2) Qiu, J.; Bai, X.; Wang, L.* *J. Phys. Chem. Lett.* **9**, 4319 (2018).
- (3) Qiu, J.; Bai, X.; Wang, L.* *J. Phys. Chem. Lett.* **10**, 637 (2019).
- (4) Wang, L.*; Qiu, J.; Bai, X.; Xu, J. *WIREs Comput. Mol. Sci.* **10**, e1435 (2020).
- (5) Xu, J.; Wang, L.* *J. Chem. Phys.* **150**, 164101 (2019).
- (6) Xiao, B.; Xu, J.; Wang, L.* DOI:10.1063/1674-0068/cjcp2006098 (2020).
- (7) Xu, J.; Wang, L.* DOI: 10.26434/chemrxiv.12820238 (2020).

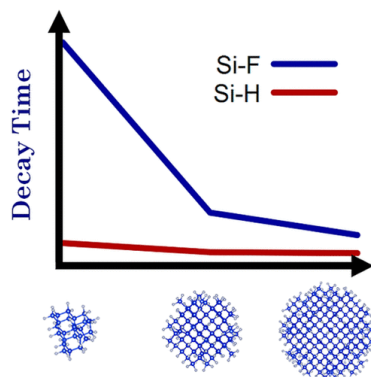
Hot Electron Cooling in Silicon Nanoclusters via Landau-Zener Non-Adiabatic Molecular Dynamics: Size Dependence and Role of Surface Termination

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Abstract In this seminar, I will describe a new express methodology for modeling excited states dynamics in large nanoscale and condensed phase systems in which the explicit computation of nonadiabatic couplings is not required. The method leverages a recent reformulation of Landau-Zener nonadiabatic transition probabilities within the computational efficient Neglect-of-Back-Reaction Approximation. Compared to popular wavefunction-based nonadiabatic molecular dynamics methods within the Neglect-of-Back-Reaction Approximation, our newly developed method is roughly an order of magnitude cheaper and costs only a little more than that of the molecular dynamics. The method is applied to modelling the non-radiative hot electron cooling in hydrogen and fluorine terminated silicon nanocrystals containing up to nearly 600 atoms. We find that hot electrons cool significantly slower in fluorine terminated silicon nanocrystals than their hydrogen terminated counterparts. The slower dynamics in the case of fluorine terminated silicon nanocrystals is rationalized by the larger energy gaps and larger mass of the fluorine atoms. We observe that the dynamics obtained using our new method yields the same qualitative picture as does the popular wavefunction based approach within the Neglect-of-Back-Reaction Approximation. Therefore, we consider the method to be a computationally efficient alternative to obtaining the nonadiabatic dynamics in large nanoscale materials, which is important to the prediction and characterization of their photophysical properties.



References

- (1) Smith, B.; Akimov, A. V. "Hot Electron Cooling in Silicon Nanoclusters via Landau-Zener Non-Adiabatic Molecular Dynamics: Size Dependence and Role of Surface Termination" *J. Phys. Chem. Lett.* **2020**, *11*, 1456-1465

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

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