VISTA Seminar

Seminar 65

March 20, 2024

10:00 am – 11:30 am EST / 3:00 – 4:30 pm GMT London / 4:00 pm – 5:30 pm CET Paris / 11 pm CST Beijing

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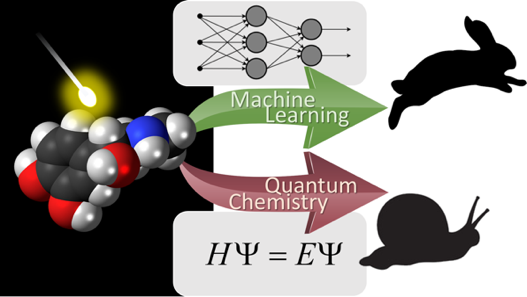
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# **Towards more accessible excited-state simulations with AI**

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Machine learning can greatly speed up quantum chemical simulations.[[1](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_1)]

I will present our ongoing journey towards making excited-state simulations more accessible with the help of AI/ML. We started it with the goal of accelerating nonadiabatic excited-state molecular dynamics which are extremely slow and resource-consuming in the on-the-fly surface-hopping framework. Our early works explored the potential gains and possible solutions for ML-accelerated nonadiabatic dynamics[[2](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_2)] and showcased the practical possibility of such simulations[[3](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_3)]. In this context, we developed an efficient approach for substantially reducing the cost of calculating excited-state properties for initial conditions which can be used to obtain precise one-photon UV/vis absorption spectra[[4](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_4)] (later, we developed an efficient universal approach for predicting related two-photon absorption spectra[[5](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_5)]). Interpolating between the initial conditions can be efficiently done with kernel methods in agreement with our systematic study of the optimal choice of ML models for single-molecule PES.[[6](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_6)] However, for dynamics requiring more data, NN approaches turn out to be a good practical choice, both in the context of developing universal Δ-learning improved semi-empirical QM method AIQM1 with a good accuracy and low cost for excited-state simulations[[7](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_7)] and for creating specialized models for nonadiabatic dynamics of specific systems[[3](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_3),[8](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_8)]. AIQM1 has been successfully applied for excited-state geometry optimizations[[7](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_7),[9](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_9)] owing to our semi-empirical baseline method[[10](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_10)] and we are currently exploring its prowess for nonadiabatic dynamics. Specialized models can be used for cost-efficient generation of the nonlinear time-resolved spectra from nonadiabatic dynamics trajectories.[[8](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_8)]

Beyond on-the-fly surface hopping dynamics, we explored ML approaches for accelerating,[[11-12](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_11)] learning trajectories as a function of time,[[13](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_13)] and learning the entire trajectories[[14](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_14)] of the quantum dissipative dynamics.

This research stimulated the development of integrated ML software ecosystems for excited-state simulations based on MLatom,[[15](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_15)] Newton-X,[[16](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_16)] and MLQD[[17](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_17)], and an increasing number of such simulations can be performed online at the <https://XACScloud.com>. General discussion about the progress in this field is given in our review[[1](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_1)] and book chapters[[18-20](file:///C:\Users\Alexey\Downloads\20240314_abstract_Dral.docx#_ENREF_18)].

**References**

[1] P. O. Dral, M. Barbatti. Molecular Excited States Through a Machine Learning Lens. *Nat. Rev. Chem.* **2021,** *5*, 388–405.

[2] P. O. Dral, M. Barbatti, W. Thiel. Nonadiabatic Excited-State Dynamics with Machine Learning. *J. Phys. Chem. Lett.* **2018,** *9*, 5660–5663.

[3] W.-K. Chen, X.-Y. Liu, W. Fang, P. O. Dral, G. Cui. Deep Learning for Nonadiabatic Excited-State Dynamics. *J. Phys. Chem. Lett.* **2018,** *9*, 6702–6708.

[4] B.-X. Xue, M. Barbatti, P. O. Dral. Machine Learning for Absorption Cross Sections. *J. Phys. Chem. A* **2020,** *124*, 7199–7210.

[5] Y. Su, Y. Dai, Y. Zeng, C. Wei, Y. Chen, F. Ge, P. Zheng, D. Zhou, P. O. Dral, C. Wang. Interpretable Machine Learning of Two‐Photon Absorption. *Adv. Sci.* **2023**, 2204902.

[6] M. Pinheiro Jr, F. Ge, N. Ferré, P. O. Dral, M. Barbatti. Choosing the right molecular machine learning potential. *Chem. Sci.* **2021,** *12*, 14396–14413.

[7] P. Zheng, R. Zubatyuk, W. Wu, O. Isayev, P. O. Dral. Artificial Intelligence-Enhanced Quantum Chemical Method with Broad Applicability. *Nat. Commun.* **2021,** *12*, 7022.

[8] S. V. Pios, M. F. Gelin, A. Ullah, P. O. Dral, L. Chen. Artificial-Intelligence-Enhanced On-the-Fly Simulation of Nonlinear Time-Resolved Spectra. *J. Phys. Chem. Lett.* **2024,** *15*, 2325–2331.

[9] T. A. Schaub, A. Zieleniewska, R. Kaur, M. Minameyer, W. Yang, C. M. Schüßlbauer, L. Zhang, M. Freiberger, L. N. Zakharov, T. Drewello, P. O. Dral, D. Guldi, R. Jasti. Tunable Macrocyclic Polyparaphenylene Nanolassos via Copper‐Free Click Chemistry. *Chem. Eur. J.* **2023,** *29*, e202300668.

[10] P. O. Dral, X. Wu, W. Thiel. Semiempirical Quantum-Chemical Methods with Orthogonalization and Dispersion Corrections. *J. Chem. Theory Comput.* **2019,** *15*, 1743–1760.

[11] A. Ullah, P. O. Dral. Speeding up quantum dissipative dynamics of open systems with kernel methods. *New J. Phys.* **2021,** *23*, 113019.

[12] L. E. Herrera Rodríguez, A. Ullah, K. J. Rueda Espinosa, P. O. Dral, A. A. Kananenka. A comparative study of diﬀerent machine learning methods for dissipative quantum dynamics. *Mach. Learn. Sci. Technol.* **2022,** *3*, 045016.

[13] A. Ullah, P. O. Dral. Predicting the future of excitation energy transfer in light-harvesting complex with artificial intelligence-based quantum dynamics. *Nat. Commun.* **2022,** *13*, 1930.

[14] A. Ullah, P. O. Dral. One-Shot Trajectory Learning of Open Quantum Systems Dynamics. *J. Phys. Chem. Lett.* **2022,** *13*, 6037–6041.

[15] P. O. Dral, F. Ge, Y.-F. Hou, P. Zheng, Y. Chen, M. Barbatti, O. Isayev, C. Wang, B.-X. Xue, M. Pinheiro Jr, Y. Su, Y. Dai, Y. Chen, S. Zhang, L. Zhang, A. Ullah, Q. Zhang, Y. Ou. MLatom 3: A Platform for Machine Learning-Enhanced Computational Chemistry Simulations and Workflows. *J. Chem. Theory Comput.* **2024,** *20*, 1193–1213. See [MLatom.com](http://MLatom.com) @ [XACScloud.com](https://XACScloud.com).

[16] M. Barbatti, M. Bondanza, R. Crespo-Otero, B. Demoulin, P. O. Dral, G. Granucci, F. Kossoski, H. Lischka, B. Mennucci, S. Mukherjee, M. Pederzoli, M. Persico, M. Pinheiro Jr, J. Pittner, F. Plasser, E. Sangiogo Gil, L. Stojanovic. Newton-X Platform: New Software Developments for Surface Hopping and Nuclear Ensembles. *J. Chem. Theory Comput.* **2022,** *18*, 6851–6865.

[17] A. Ullah, P. O. Dral. MLQD: A package for machine learning-based quantum dissipative dynamics. *Comput. Phys. Commun.* **2024,** *294*, 108940.

[18] L. Zhang, A. Ullah, M. Pinheiro Jr, M. Barbatti, P. O. Dral, Excited-state dynamics with machine learning. In *Quantum Chemistry in the Age of Machine Learning*, Dral, P. O., Ed. Elsevier: Amsterdam, Netherlands, **2023**; pp. 329–353.

[19] J. Westermayr, P. O. Dral, P. Marquetand, Learning excited-state properties. In *Quantum Chemistry in the Age of Machine Learning*, Dral, P. O., Ed. Elsevier: Amsterdam, Netherlands, **2023**; pp. 467–488.

[20] J. Li, M. Vacher, P. O. Dral, S. A. Lopez, Machine learning methods in photochemistry and photophysics. In *Theoretical and Computational Photochemistry: Fundamentals, Methods, Applications and Synergy with Experimentation*, García-Iriepa, C.; Marazzi, M., Eds. Elsevier: **2023**; pp. 163–189.

**Implementation of energy and gradient for the TDDFT- approximate auxiliary function (aas) method**

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A graph of excitation energy

Description automatically generatedA person smiling with a backpack

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Gold and silver nanoparticles can exhibit unique physical and chemical properties such as plasmonic resonances. These types of plasmonic nanoparticles are usually large, which leads to difficulty in computing their optical properties using the TDDFT method. In this work, we have implemented the TDDFT-aas method, which is an approximate TDDFT method. In this method, instead of calculating the exact two-center electron integrals in the *K* coupling matrix when solving the Casida equation, we approximate the integrals, thereby reducing the computational cost. Compared with the related TDDFT+TB method, a new type of gamma function is used when calculating the *K* coupling matrix, which does not depend on the tight binding parameters. The calculated absorption spectra of silver and gold nanoparticles using TDDFT-aas show good agreement with TDDFT+TB results. In addition, we have also implemented the analytical excited-state gradients for the TDDFT-aas method, which makes it possible to calculate the emission energy of the systems.

**How to connect**

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 65

Time: Mar 20, 2024 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting

<https://buffalo.zoom.us/j/98974911660?pwd=QlBIYWwzWVFtWFBPMi92OXpCekhIZz09>

Meeting ID: 989 7491 1660

Passcode: 423953