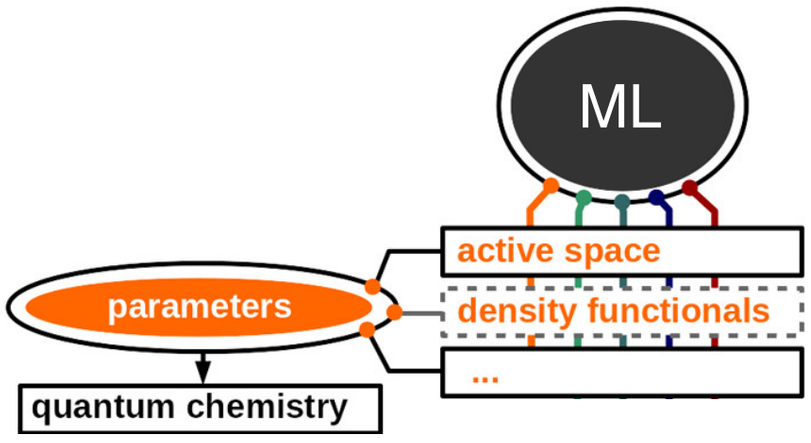
Machine learning to analyse data from ab initio molecular dynamics simulations

Morgane VACHER

CEISAM
Chimie et Interdisciplinarité
Synthèse, Analyse,
Modélisation

cnrs

Machine learning & quantum chemistry



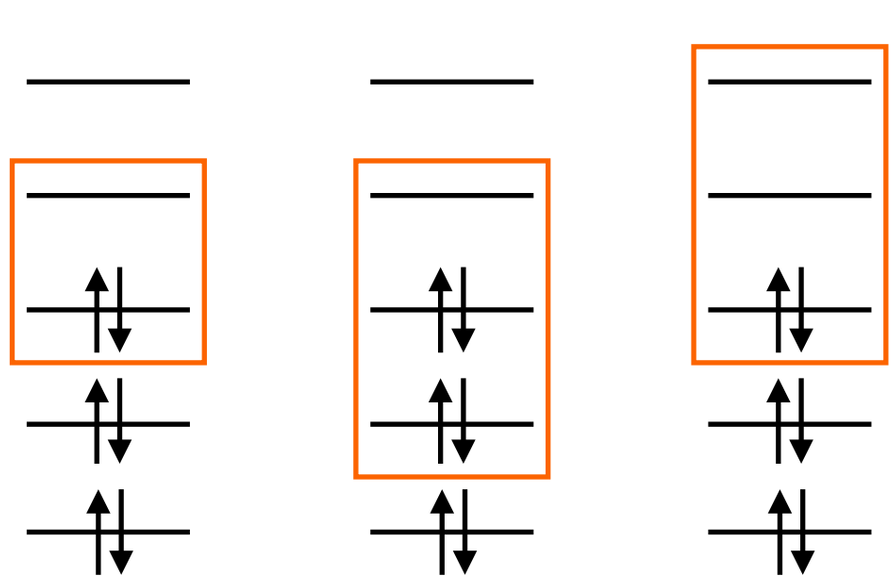
JCTC Journal of Chemical Theory and Computation

pubs.acs.org/JCTC Article

Automation of Active Space Selection for Multireference Methods via Machine Learning on Chemical Bond Dissociation

WooSeok Jeong,‖ Samuel J. Stoneburner,‖ Daniel King, Ruye Li, Andrew Walker, Roland Lindh, and Laura Gagliardi*

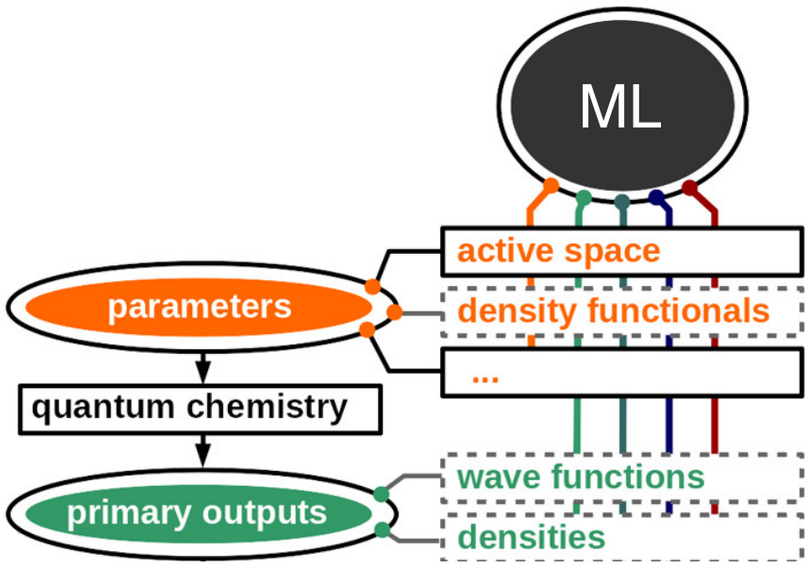
Cite This: *J. Chem. Theory Comput.* 2020, 16, 2389–2399 Read Online



What active space to choose?

Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

Machine learning & quantum chemistry



JCTC Journal of Chemical Theory and Computation

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Artificial Neural Networks Applied as Molecular Wave Function Solvers

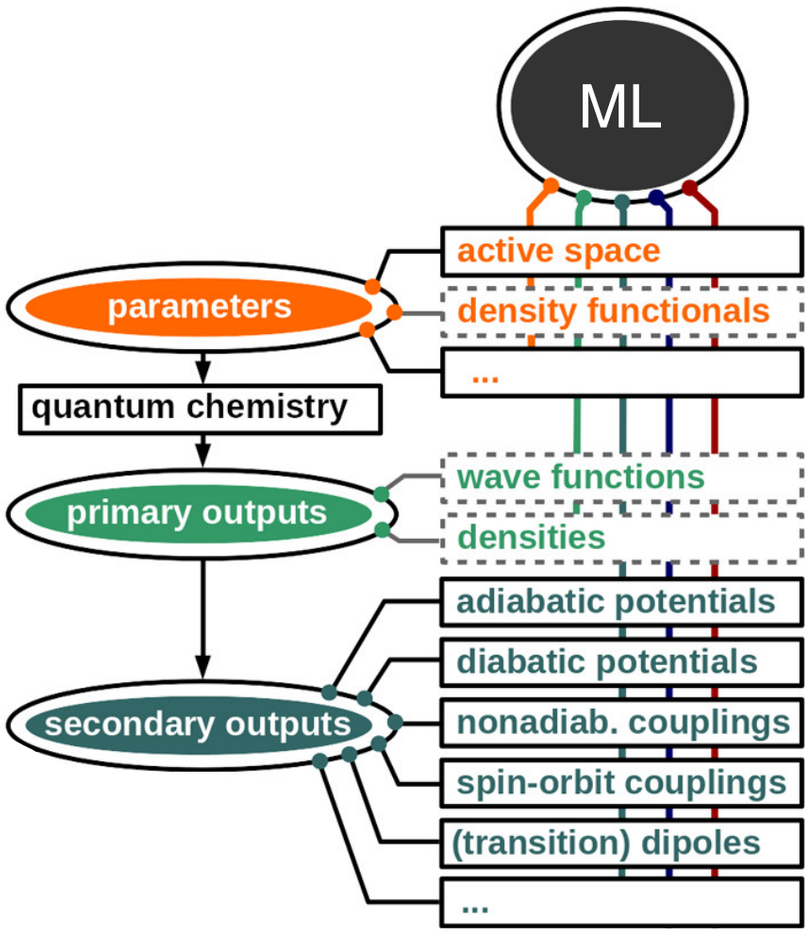
Peng-Jian Yang,* Mahito Sugiyama,* Koji Tsuda,* and Takeshi Yanai*

Cite This: *J. Chem. Theory Comput.* 2020, 16, 3513–3529 Read Online

$$\Psi = \sum_{v} C_v \Psi_v \ ?$$

Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

Machine learning & quantum chemistry



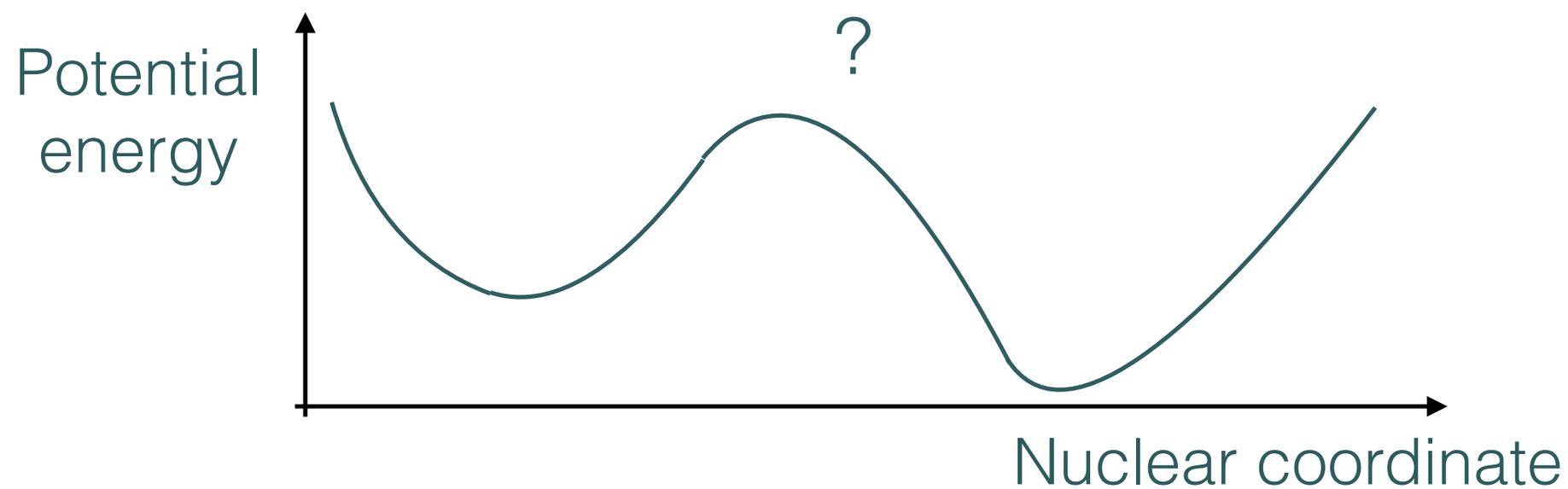
Hierarchical machine learning of potential energy surfaces

Cite as: J. Chem. Phys. **152**, 204110 (2020); https://doi.org/10.1063/5.0006498
Submitted: 04 March 2020 . Accepted: 06 May 2020 . Published Online: 27 May 2020

Pavlo O. Dral, Alec Owens, Alexey Dral, and Gábor Csányi

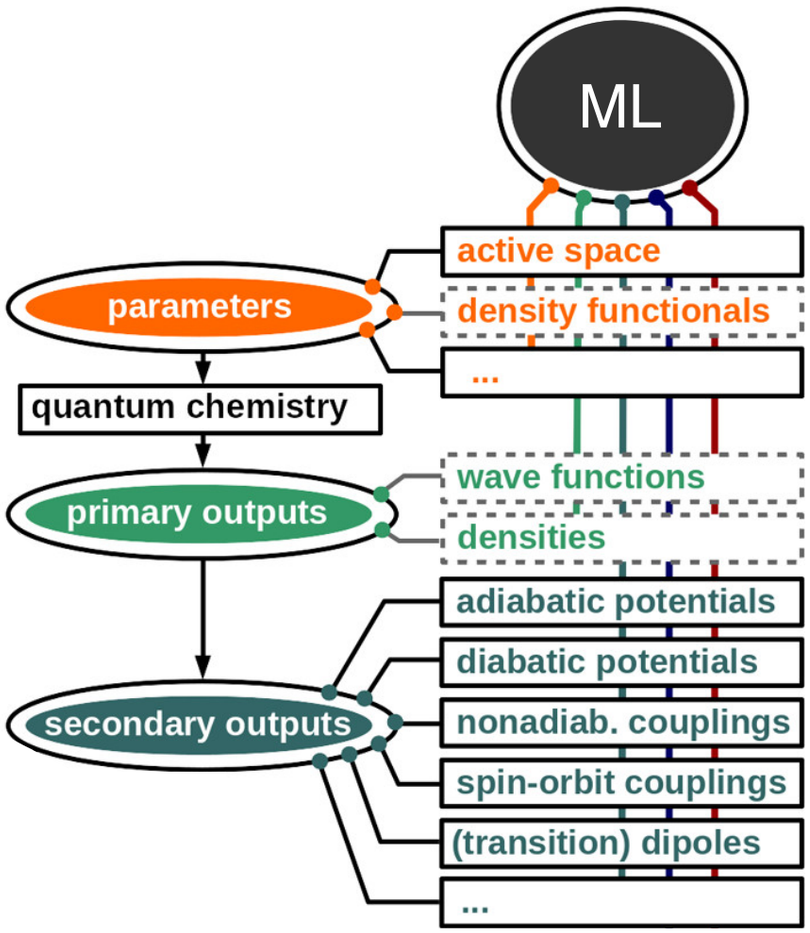
COLLECTIONS

Paper published as part of the special topic on Machine Learning Meets Chemical Physics
Note: This paper is part of the JCP Special Topic on Machine Learning Meets Chemical Physics.



Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

Machine learning & quantum chemistry



The Journal of Chemical Physics

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The Journal of Physical Chemistry Letters

ACS AuthorChoice
Letter

Cite This: J. Phys. Chem. Lett. 2018, 9, 5660−5663

pubs.acs.org/JPCL

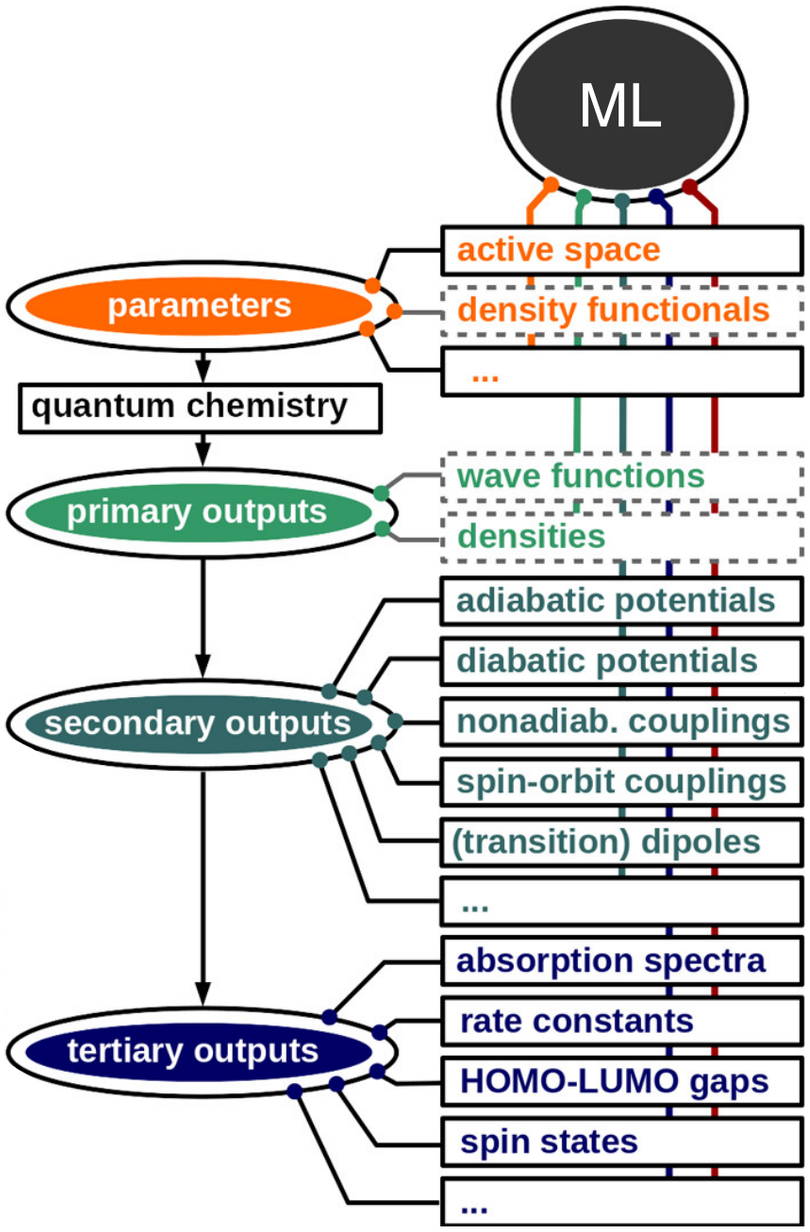
Nonadiabatic Excited-State Dynamics with Machine Learning

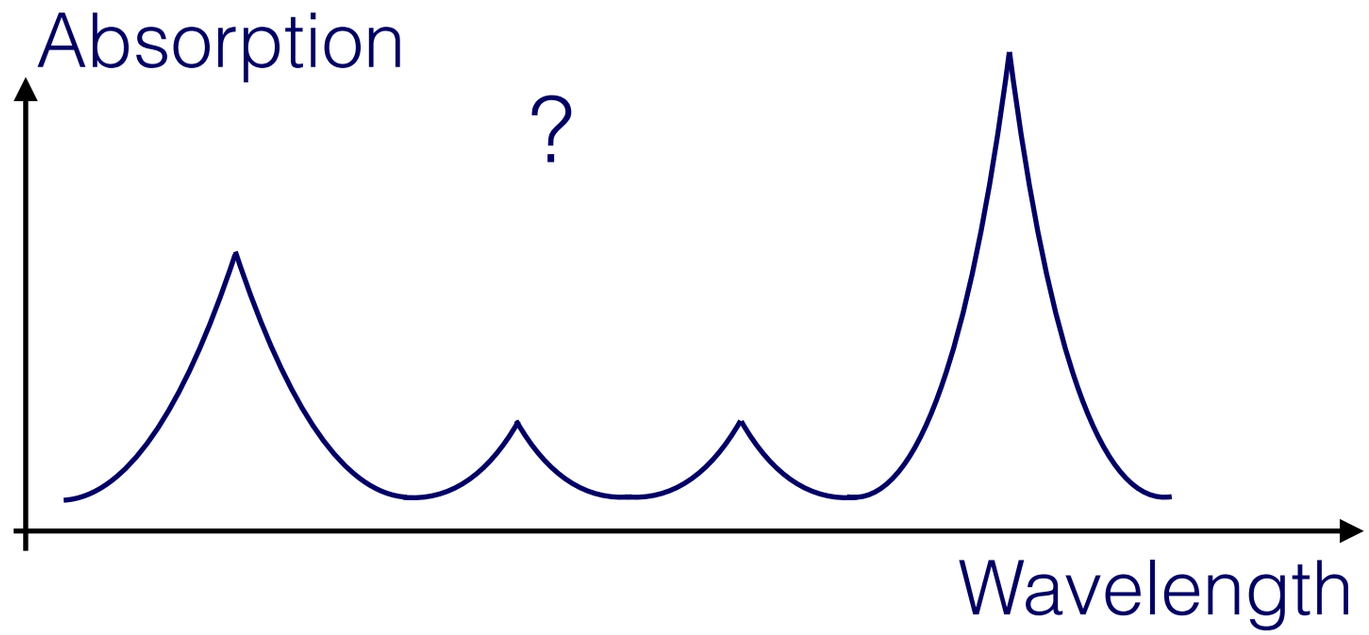
Pavlo O. Dral,*,† Mario Barbatti,*,‡ and Walter Thiel*,†

†Max-Planck-Institut für Kohlenforschung, Kaiser-Wilhelm-Platz 1, 45470 Mülheim an der Ruhr, Germany
‡Aix Marseille Univ, CNRS, ICR, Marseille, France

Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

Machine learning & quantum chemistry





THE JOURNAL OF PHYSICAL CHEMISTRY A

pubs.acs.org/JPCA Article

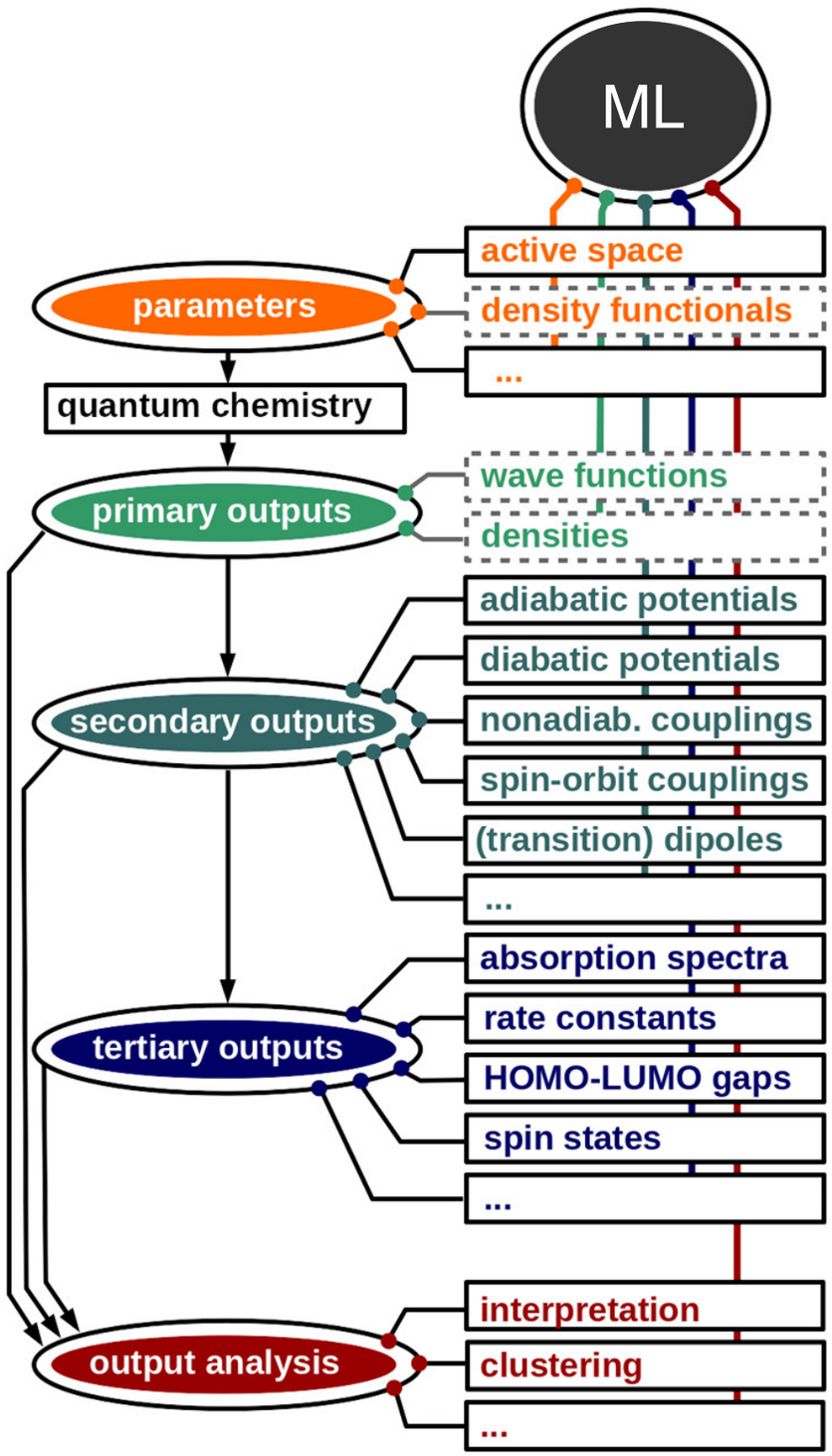
A Deep Neural Network for the Rapid Prediction of X-ray Absorption Spectra

C. D. Rankine, M. M. M. Madkhali, and T. J. Penfold*

Cite This: *J. Phys. Chem. A* 2020, 124, 4263–4270 Read Online

Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

Machine learning & quantum chemistry



Can we use **machine learning** algorithms to help the interpretation of AIMD simulations and to extract physical insights?

Chemical Science

EDGE ARTICLE

Check for updates

Cite this: *Chem. Sci.*, 2019, **10**, 2298

All publication charges for this article have been paid for by the Royal Society of Chemistry

How machine learning can assist the interpretation of *ab initio* molecular dynamics simulations and conceptual understanding of chemistry†

Florian Häse, [a] Ignacio Fdez. Galván, [b] Alán Aspuru-Guzik, [cde] Roland Lindh [b] and Morgane Vacher *[b]

ICPEAC2019 IOP Publishing

Journal of Physics: Conference Series **1412** (2020) 042003 doi:10.1088/1742-6596/1412/4/042003

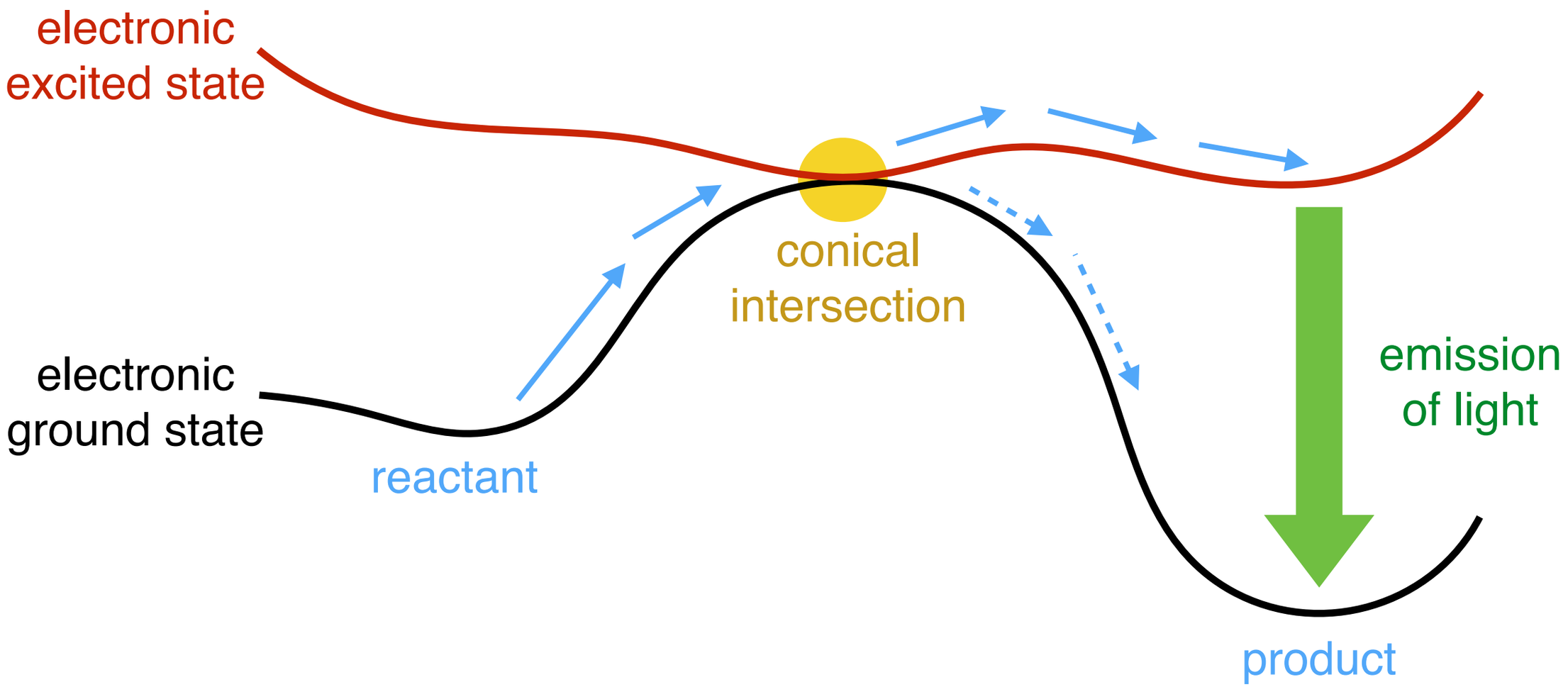
Machine learning for analysing ab initio molecular dynamics simulations

Florian Häse,[1,2,3,4] Ignacio Fdez. Galván,[5] Alán Aspuru-Guzik,[2,3,4,6] Roland Lindh[5] and Morgane Vacher[7]

Westermayr and Marquetand, *Chem. Rev.* **121**, 9873-9926 (2021)

What is chemiluminescence?

Definition: Emission of light (luminescence) as the result of a chemical reaction



Chemiexcitation

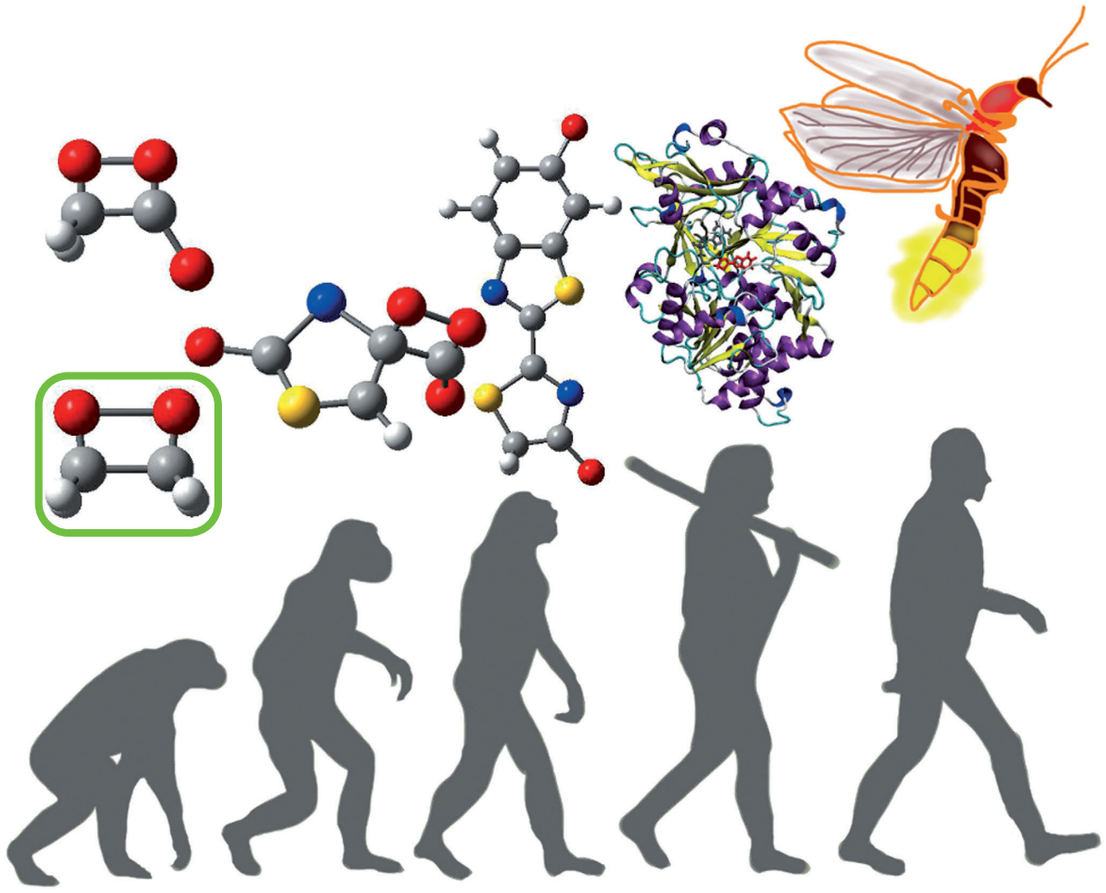
Population of an excited state as the result of a chemical reaction

What is chemiluminescence?

➜ Communication to attract partners, hunting to lure preys, defence to avoid predators

➜ In vivo imaging in medicine, biosensing for environmental polluants, food industry, etc.

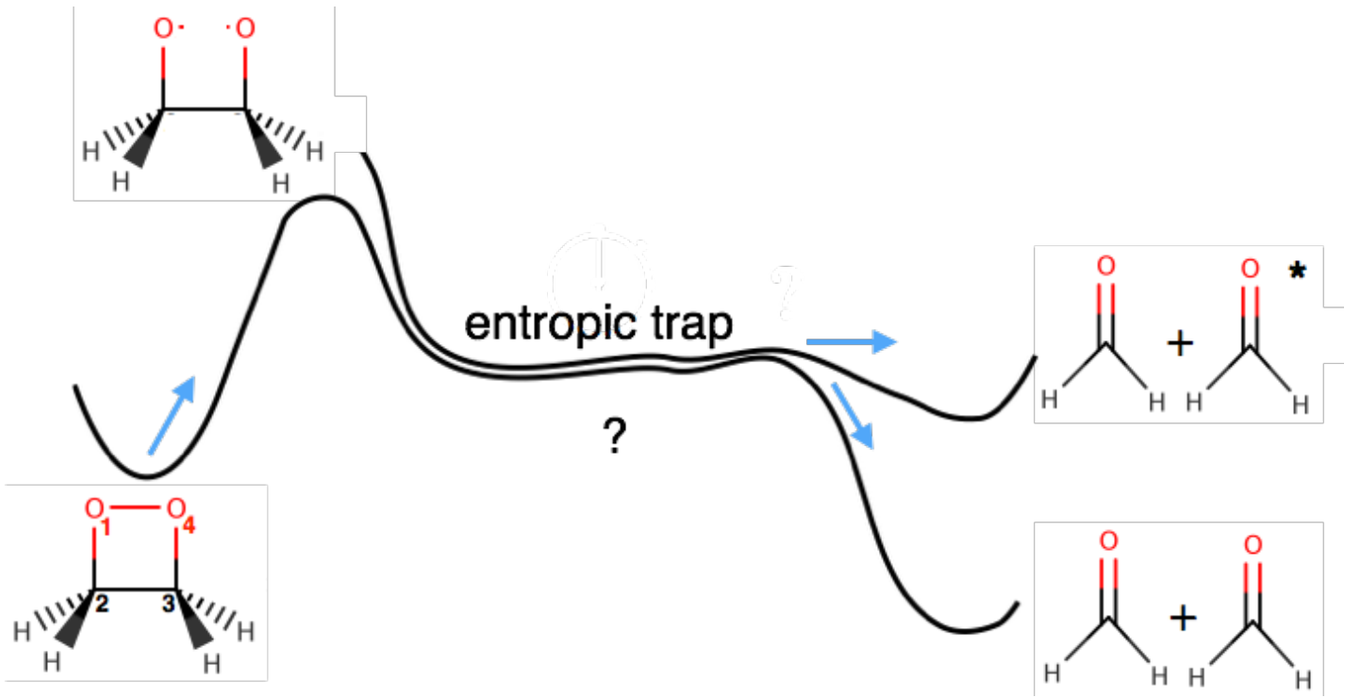
Structures of model compounds

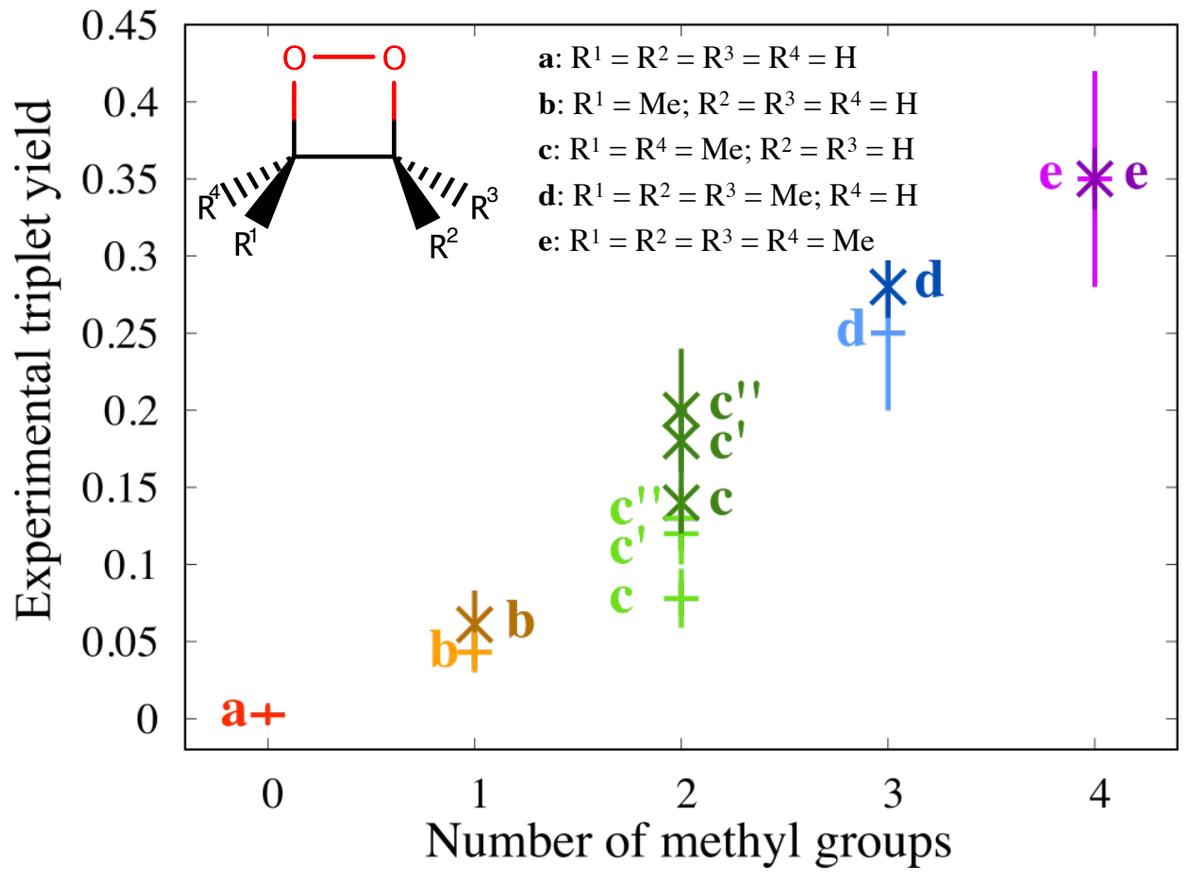


Vacher et al, *Chem. Rev.* **118**, 6927-6974 (2018)

Chemiexcitation in 1,2-dioxetane

Decomposition into fundamental or excited formaldehyde molecules





➜ Yield of triplet excited states > yield of singlet excited sates ✔

➜ Increase in the chemiexcitation yield upon methyl substitution ?

Vacher et al, *J. Phys. Chem. Letters*, **8** , 3790-3794 (2017)

Adam et al, *J. Am. Chem. Soc.*, **107**,410-416 (1985)

Theoretical approach

Ab initio molecular dynamics simulations

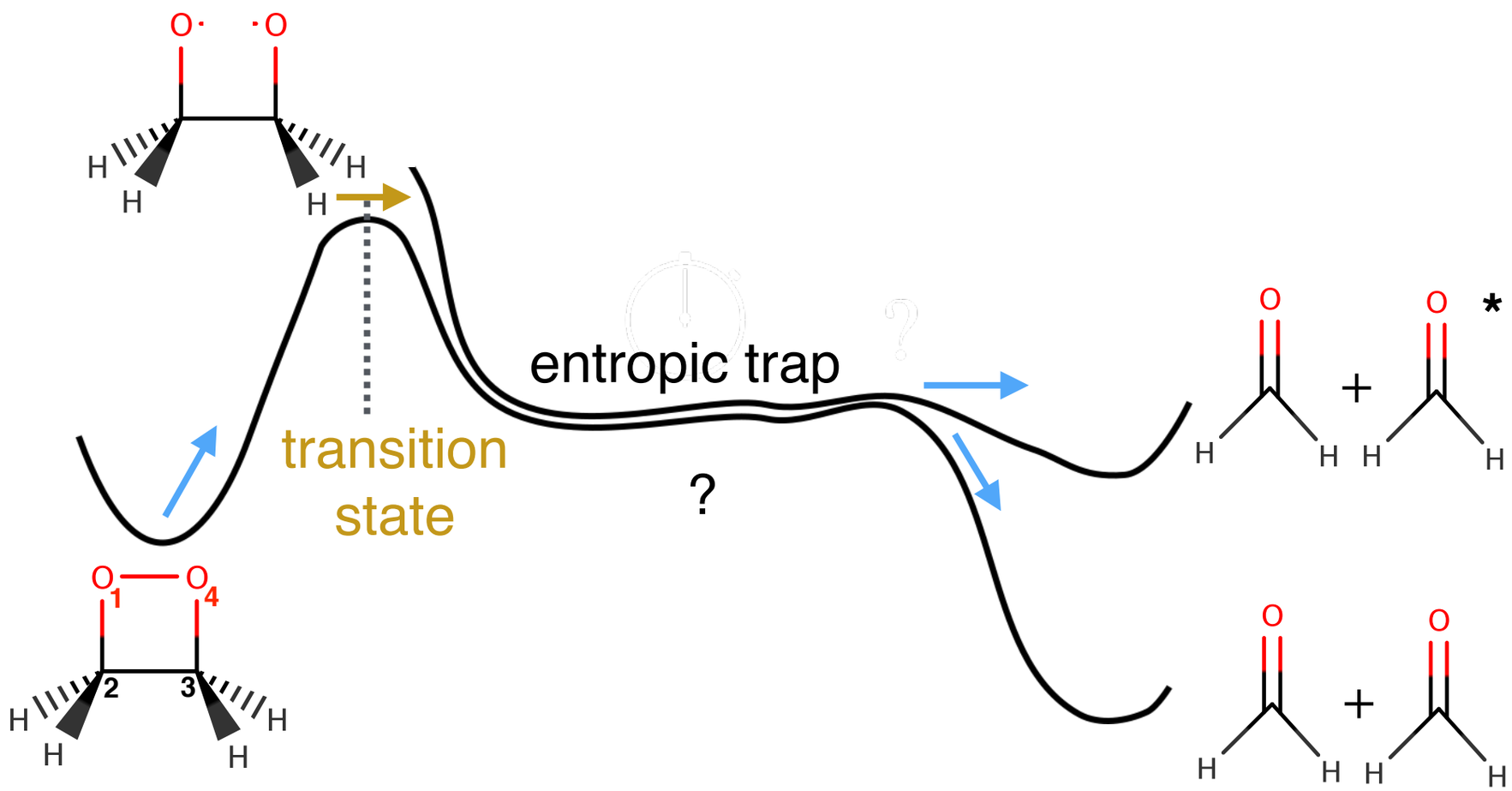
- Born-Oppenheimer dynamics
- non-adiabatic dynamics (surface hopping) including 4 singlet states

Electronic structure method

- CASSCF(12-in-10)
- ANO-RCC-VTZP basis set

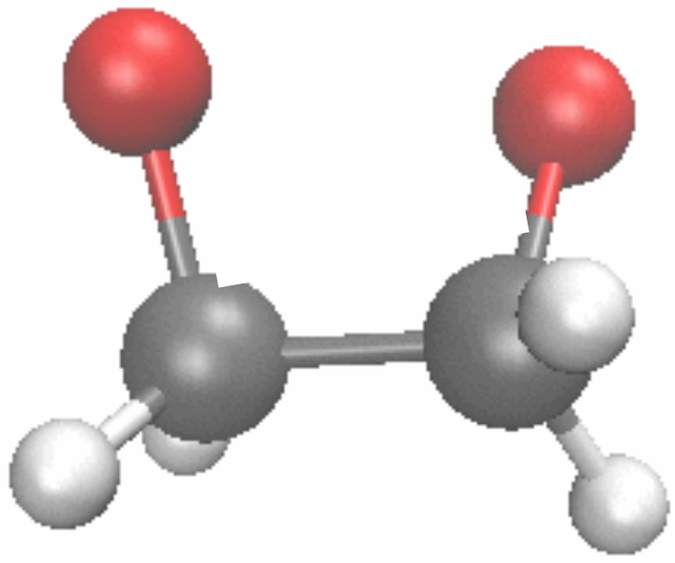
Initial conditions

- Transition state geometry with 1kcal/mol kinetic energy in the forward direction
- 150 trajectories sampled from the Wigner distribution (Newton-X package)



Vacher et al, *J. Chem. Theory Comput.*, **13** (6), 2448-2457 (2017)

Dissociation of 1,2-dioxetane



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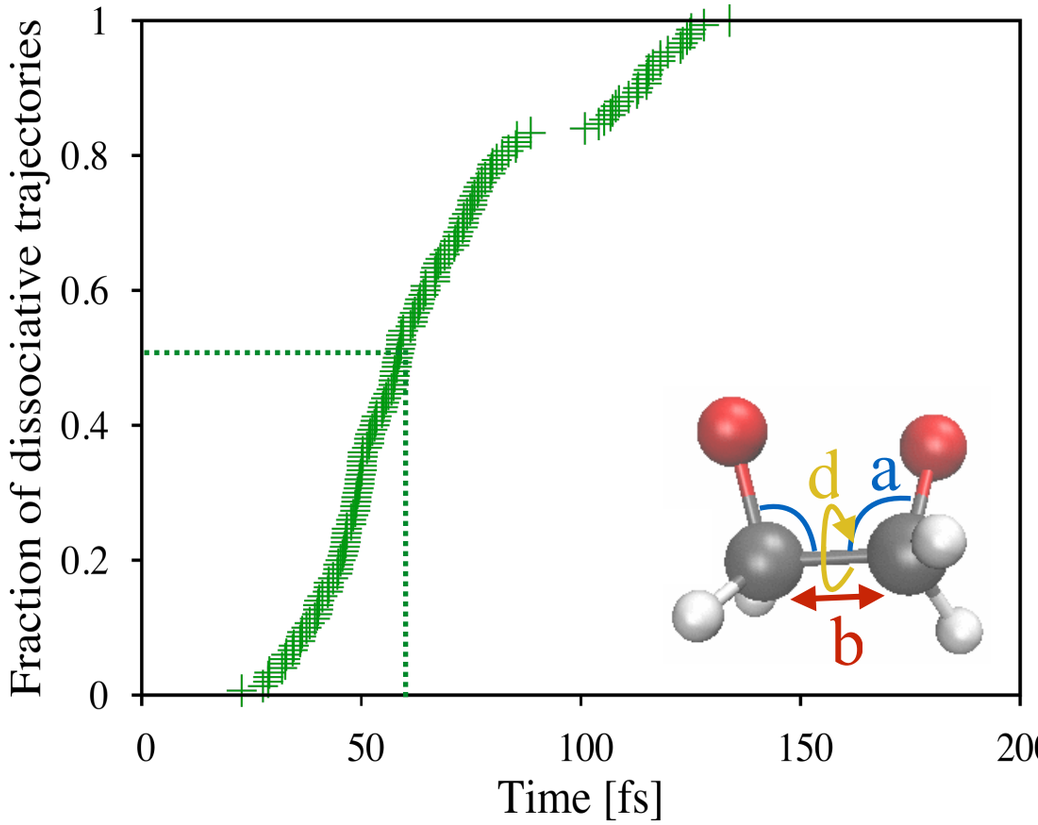
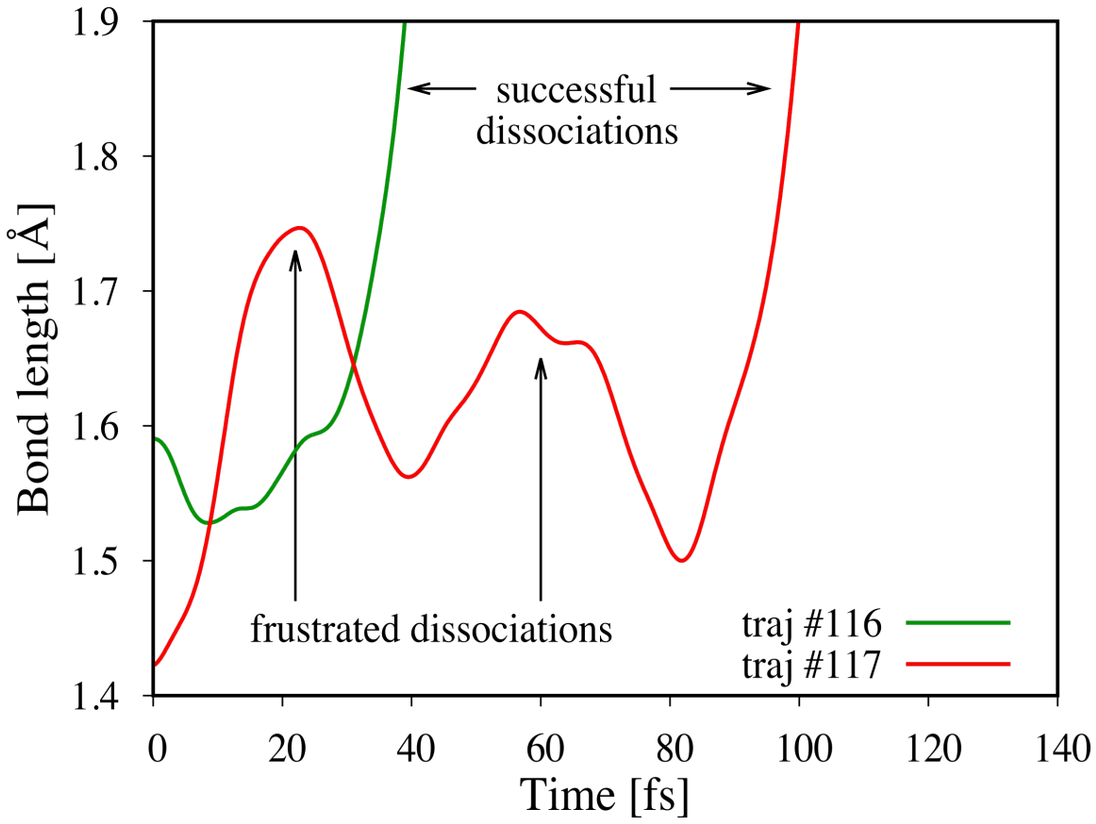
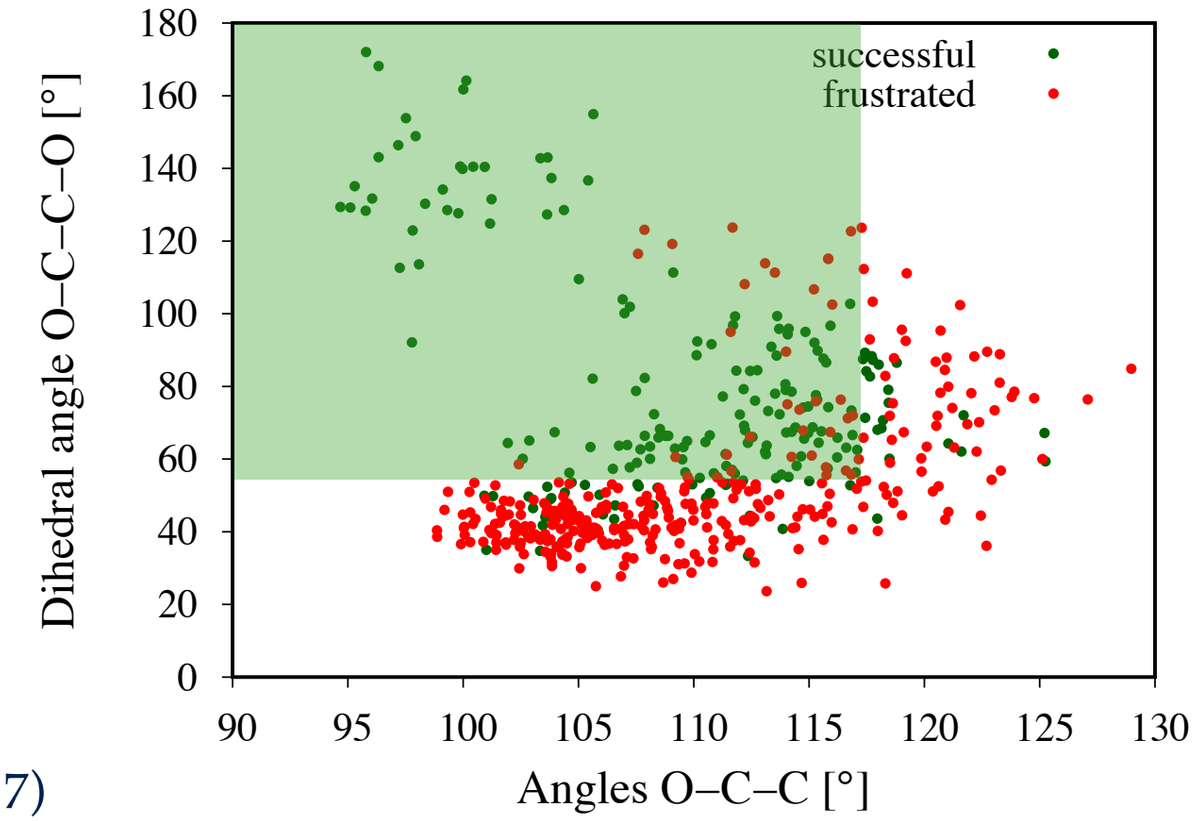




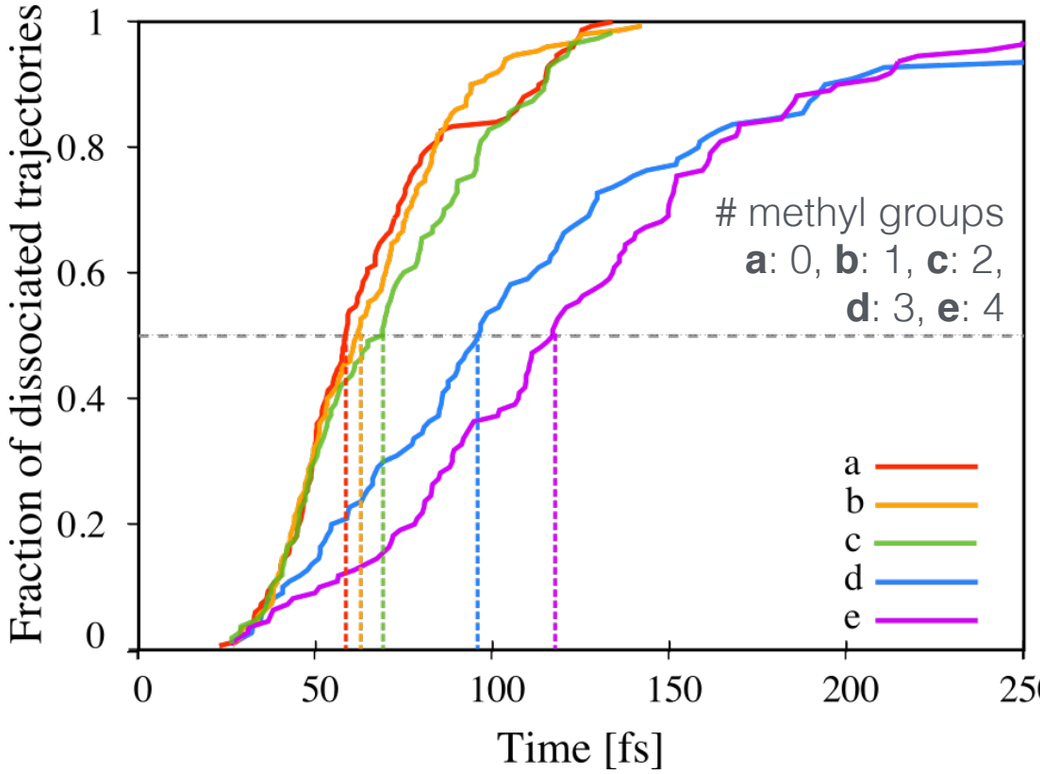
➜ Dissociation time scale
between t = 25 fs and t = 140 fs
half-time of 59 fs

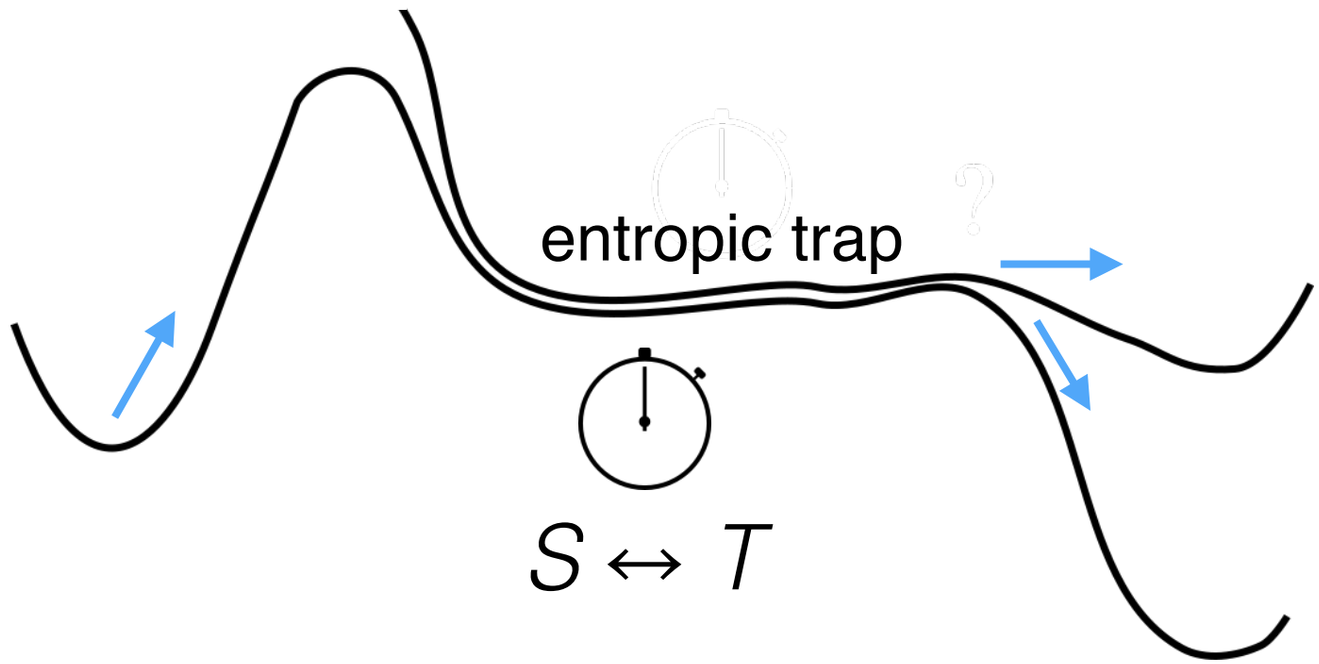
➜ Geometrical conditions necessary
O-C-C-O dihedral > 55°
O-C-C angle < 117°



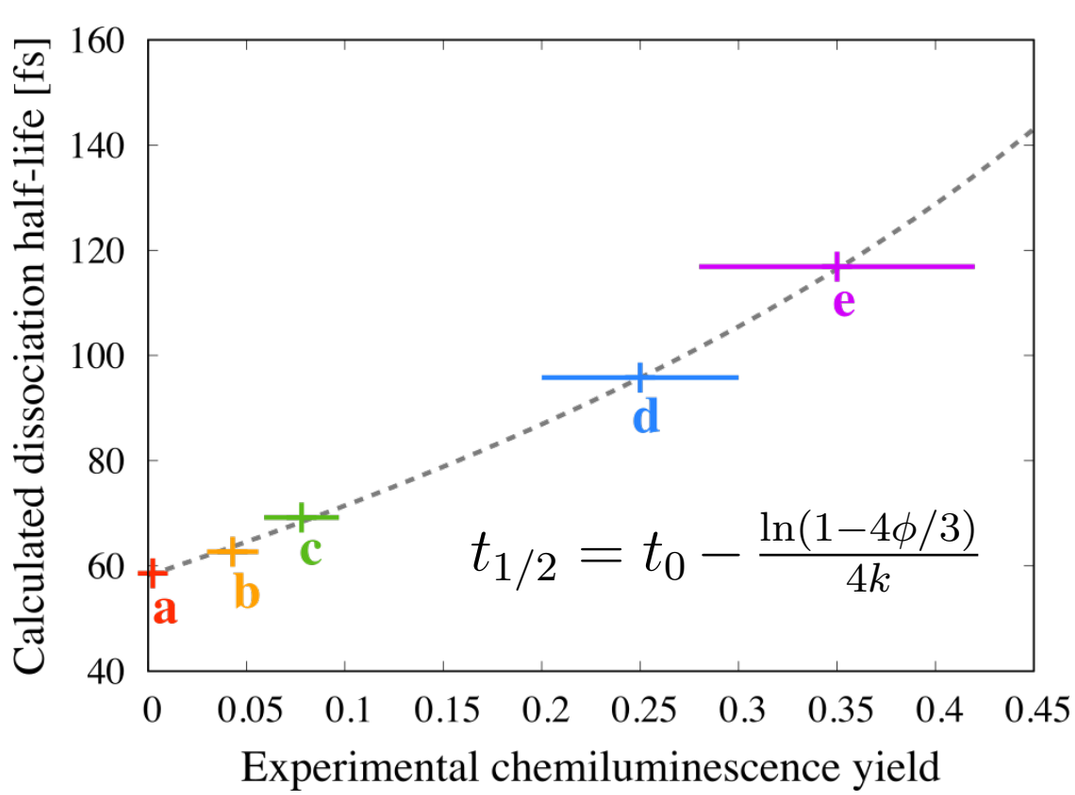
Vacher et al, *J. Chem. Theory Comput.*, **13** (6), 2448-2457 (2017)

Effect of methyl substitution





➜ The longer the system stays in the entropic trap, the more population is transferred from *S* to *T* and the higher the chemiexcitation yield is.



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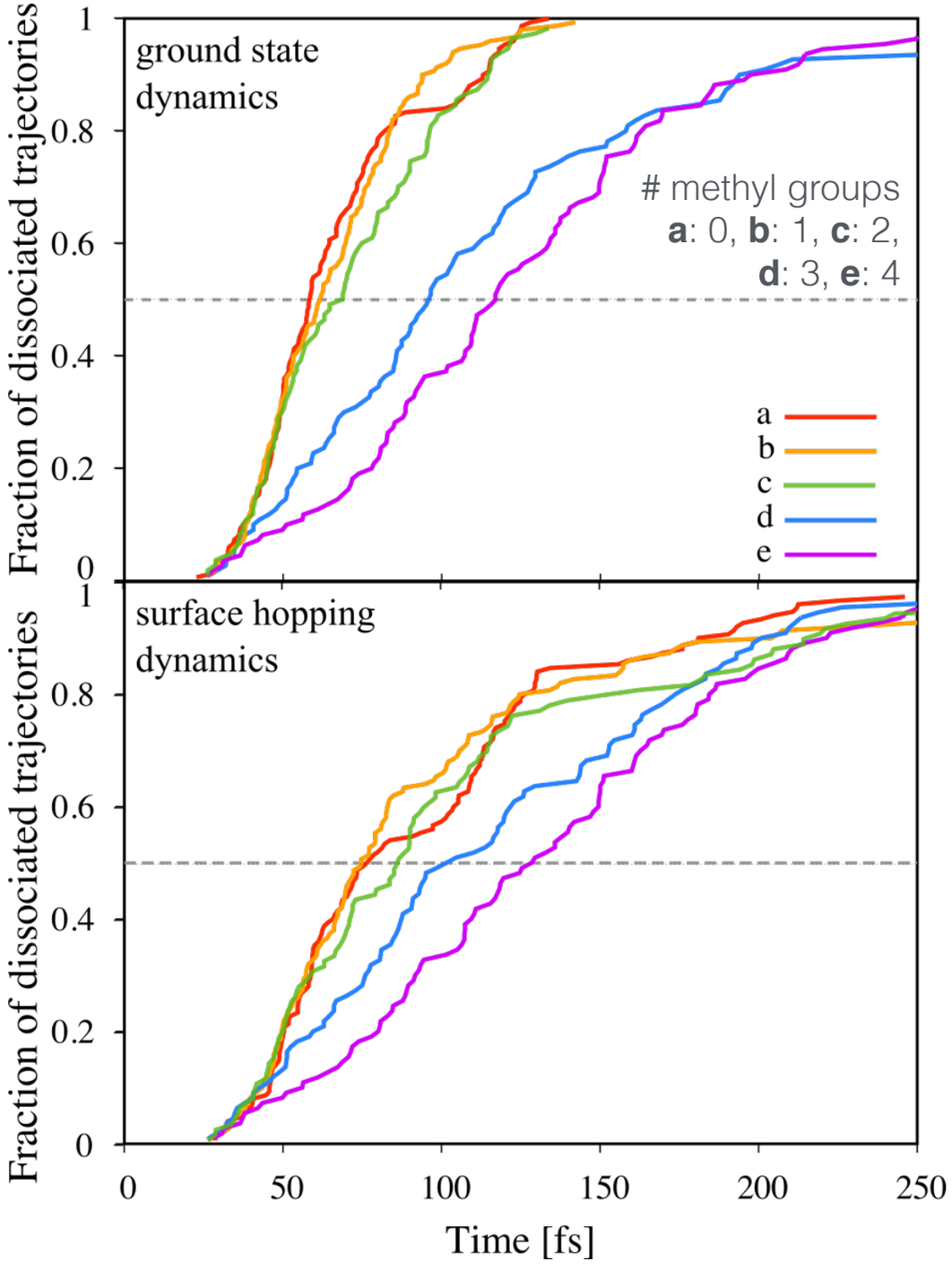
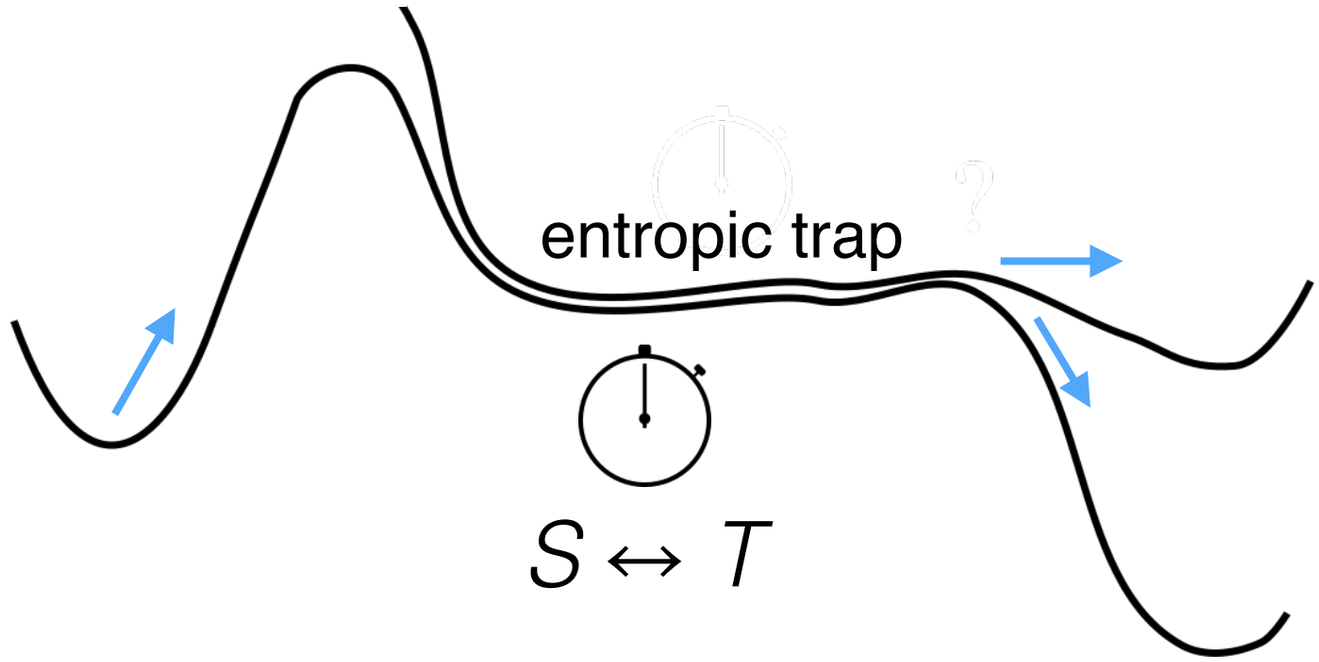
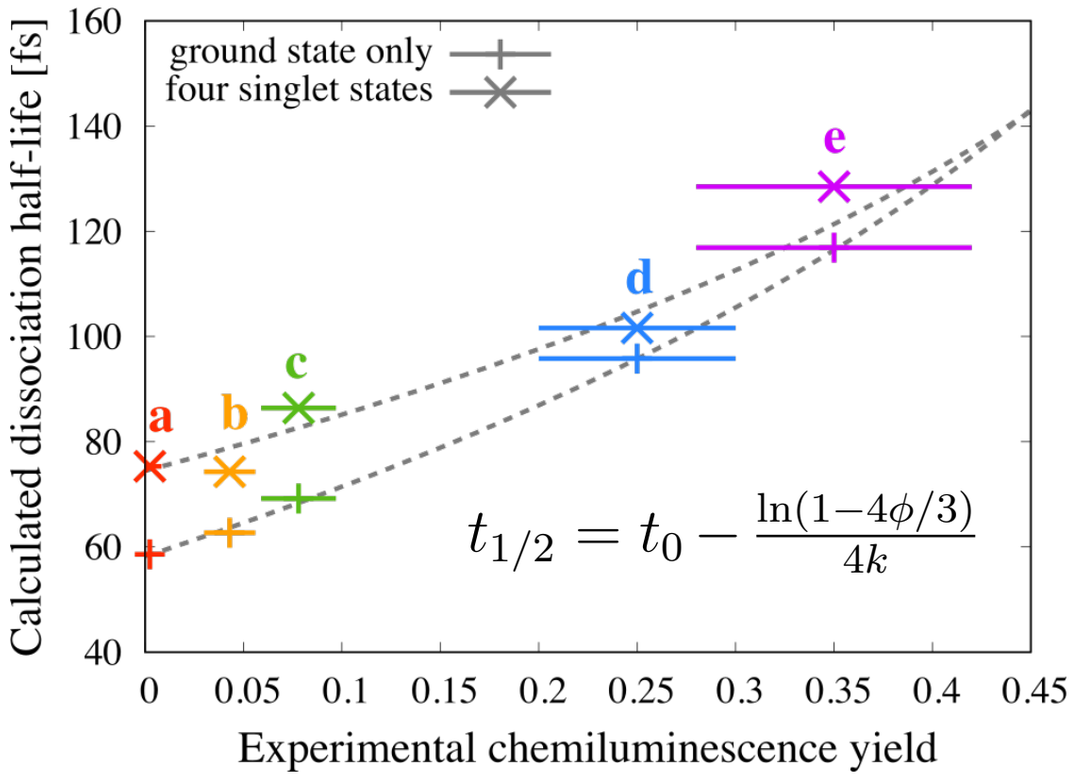




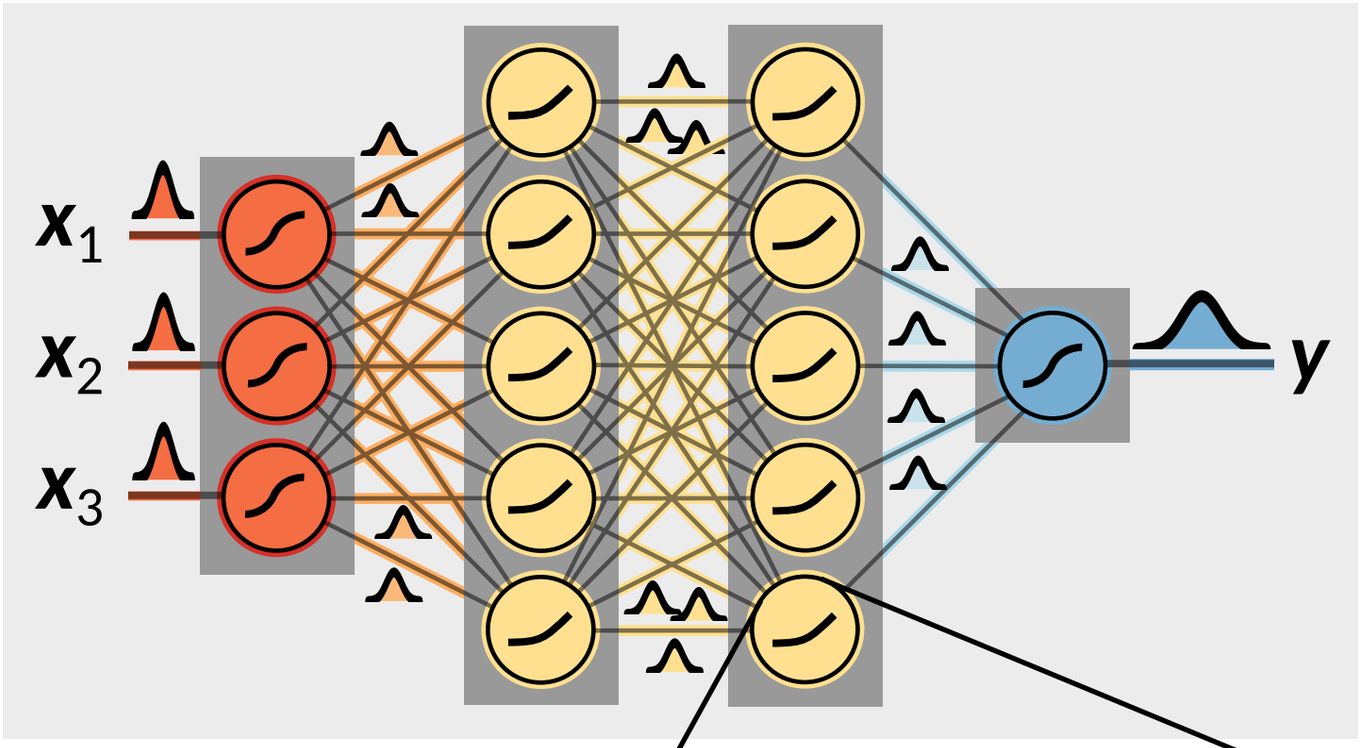


Vacher et al, *J. Phys. Chem. Letters*, **8** , 3790-3794 (2017)

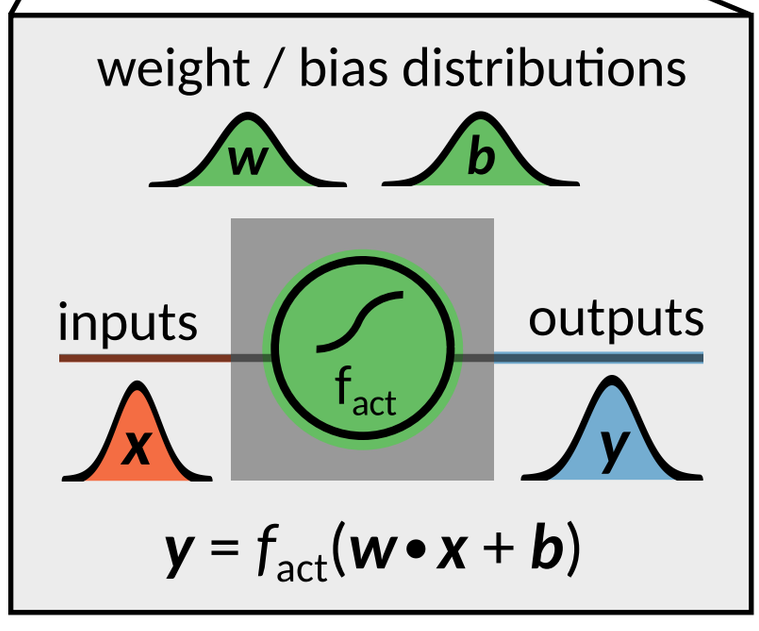
Prediction of dissociation time

BNN1:
initial nuclear
geometry

BNN2:
+ nuclear
velocities

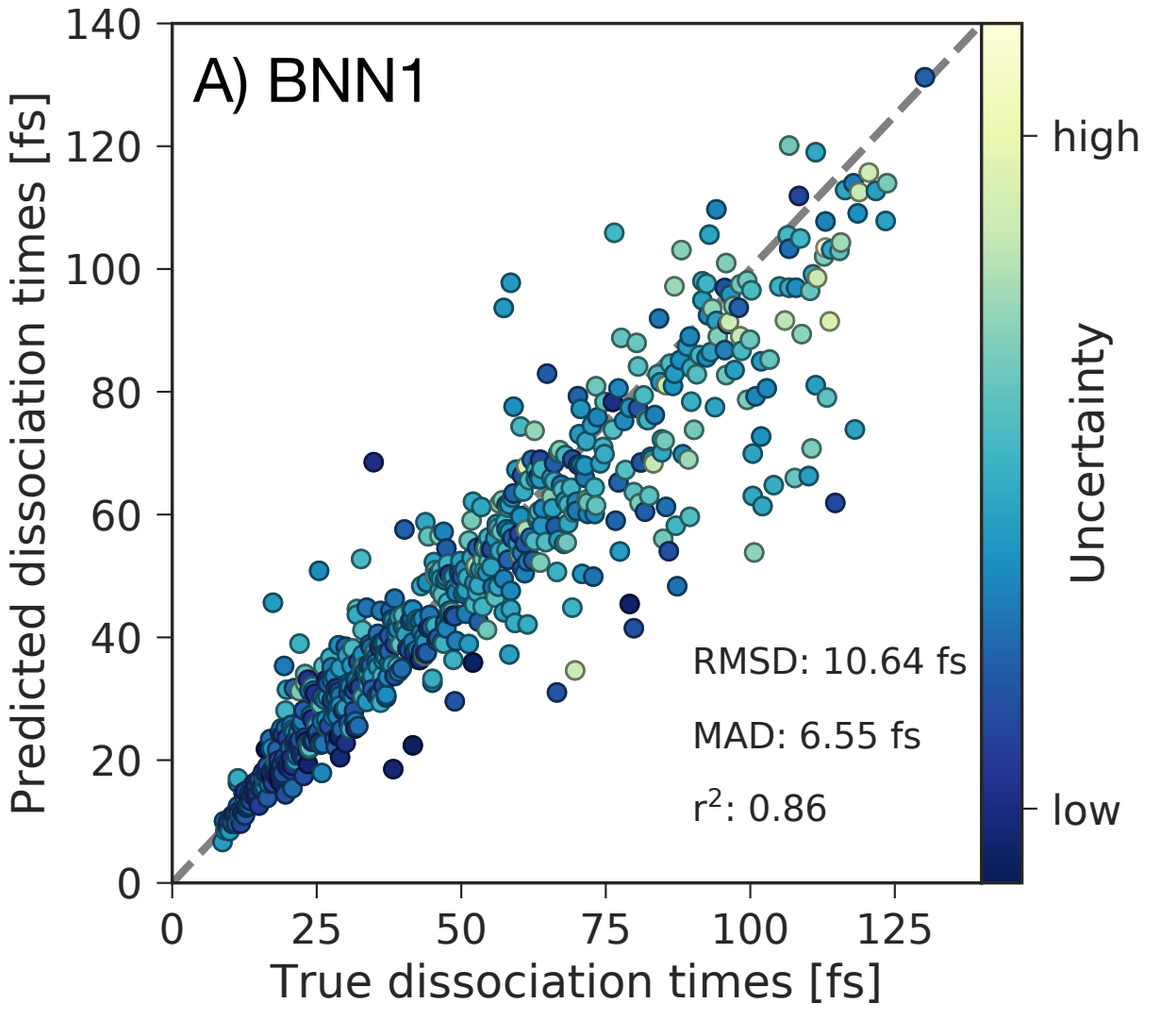


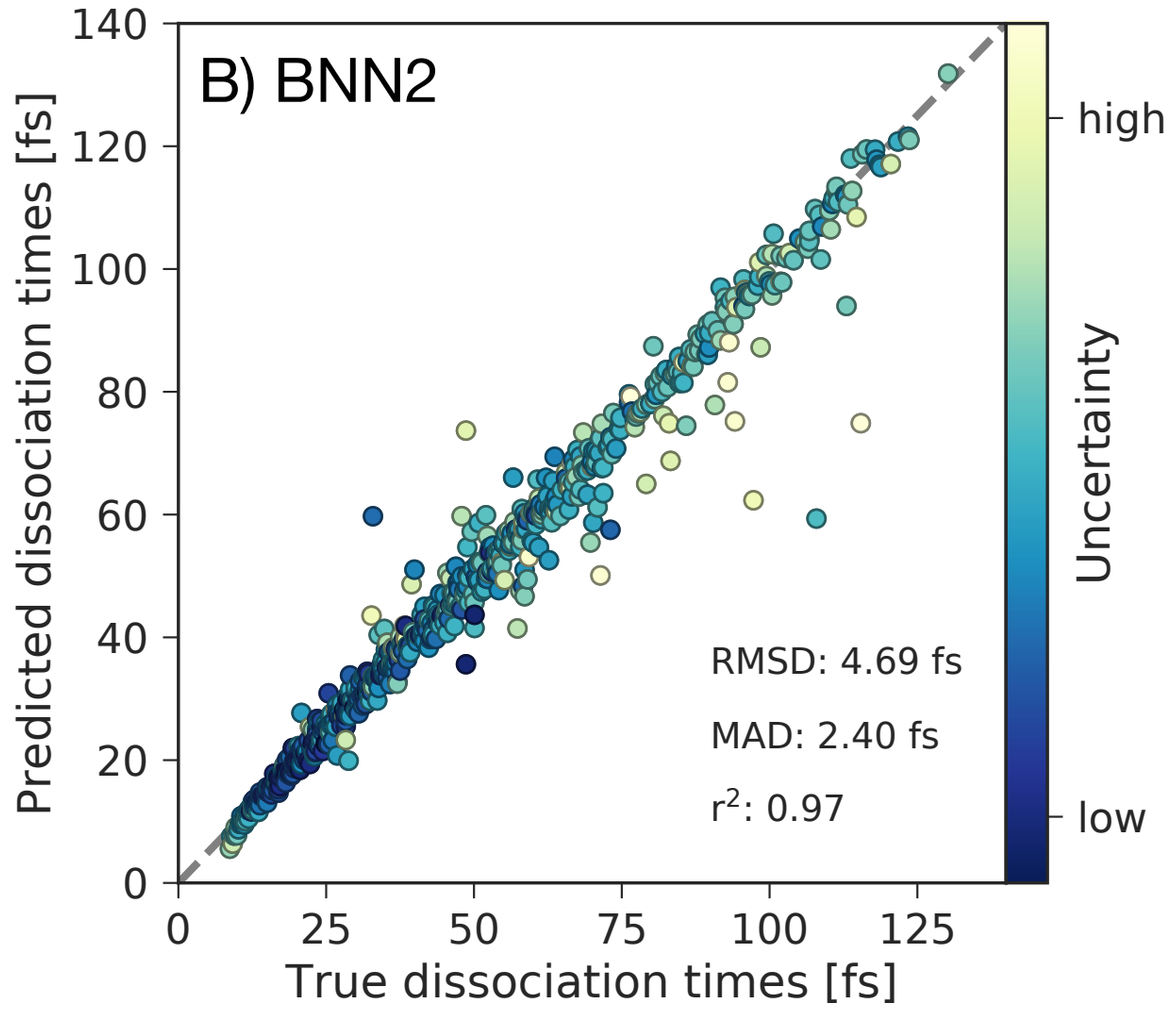
4 layers of 130 neurons
Activation function: Leaky ReLU



Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *Chem. Science*, **10**, 2298-2307 (2019)

Prediction of dissociation time

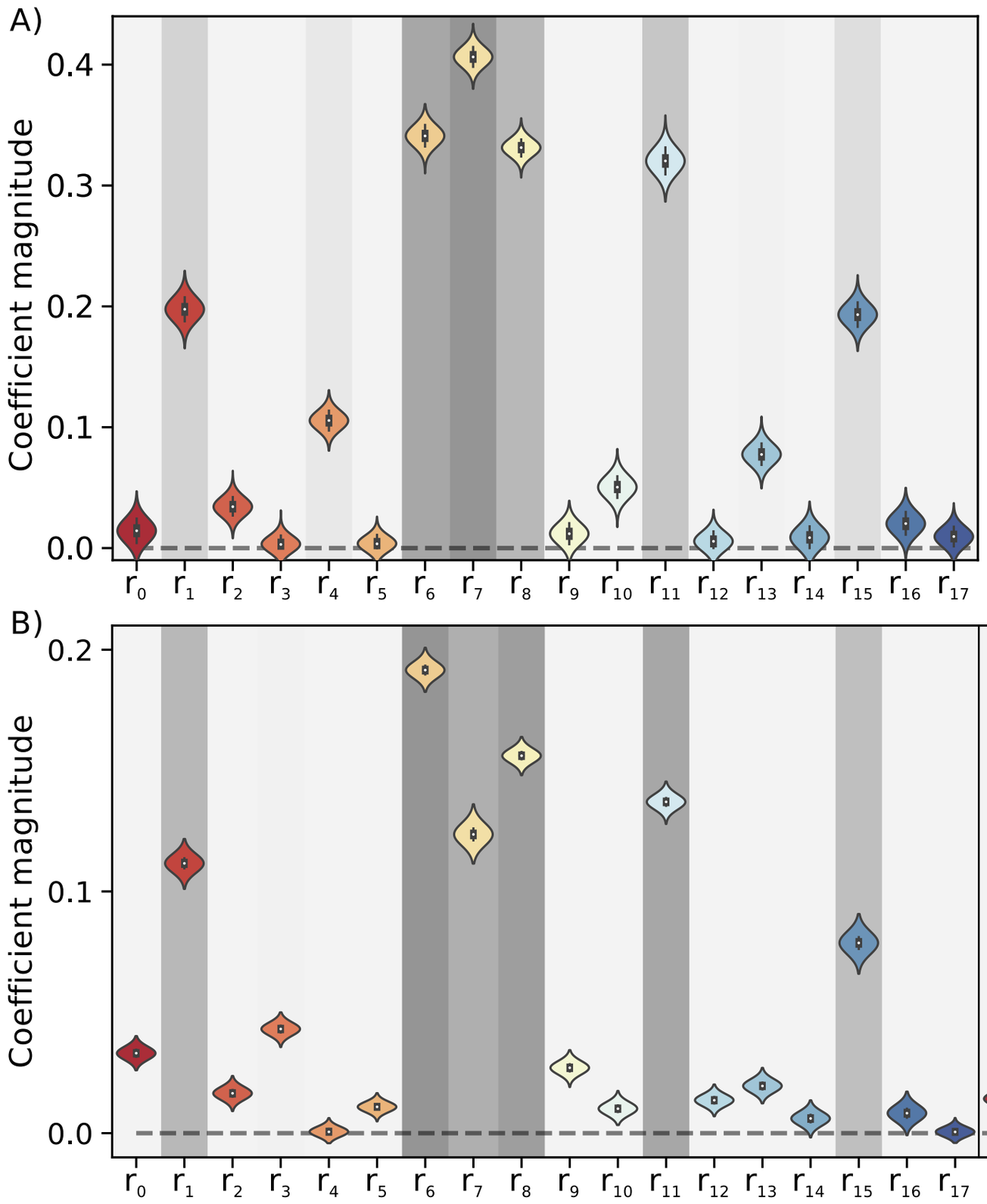


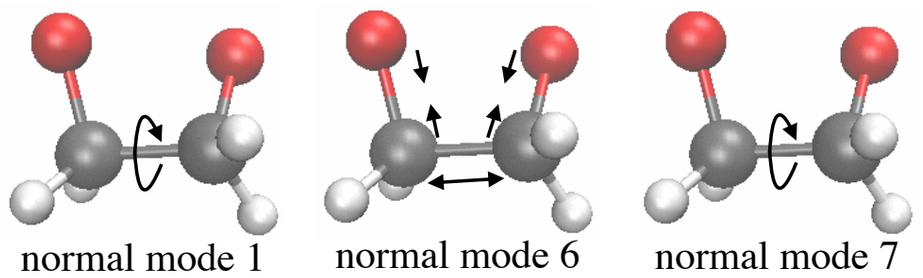


➜ Accurate predictions of the dissociation times of 1,2-dioxetane

Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *Chem. Science*, **10**, 2298-2307 (2019)

Analysis of the trained BNN 1

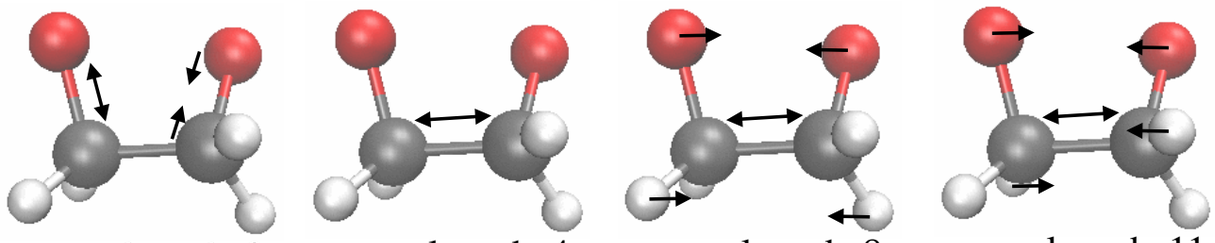




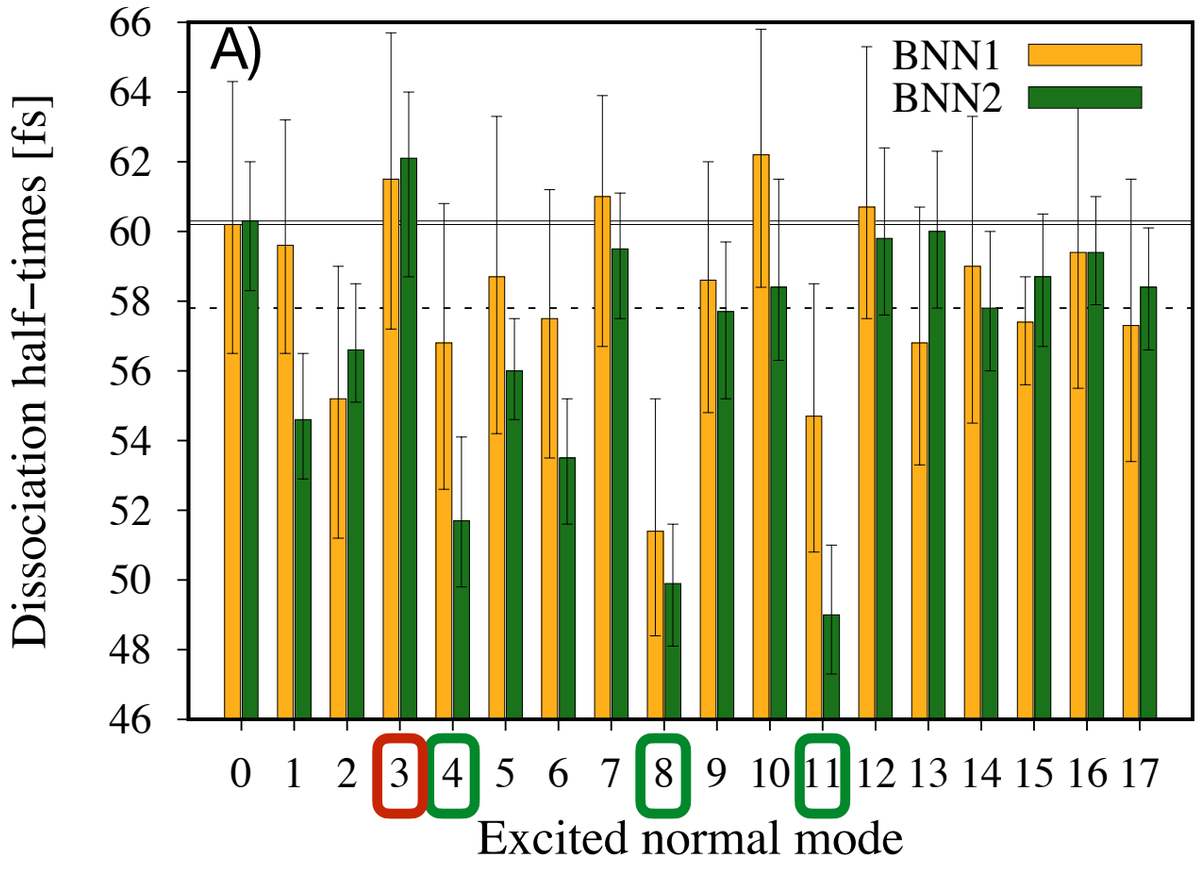
Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *Chem. Science*, **10**, 2298-2307 (2019)

Analysis of the trained BNN 2

- Predictions of dissociation times for vibrational excited states



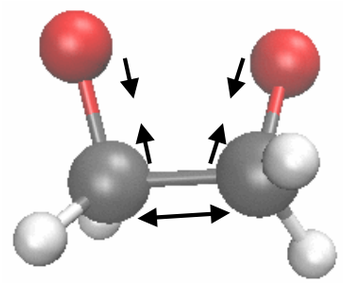
Ensemble	8	11	4	3
N_{frus}	1.08	1.17	1.18	1.52



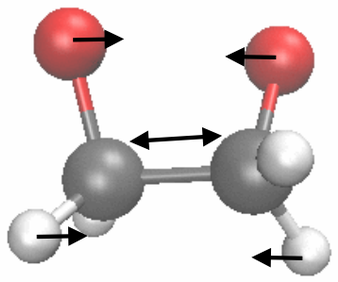
Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *Chem. Science*, **10**, 2298-2307 (2019)

Interpretation of the trained BNN

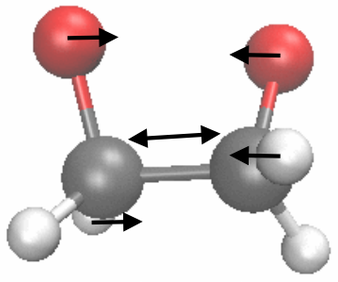
➜ Correlation between nuclear coordinates and dissociation times



normal mode 6



normal mode 8



normal mode 11

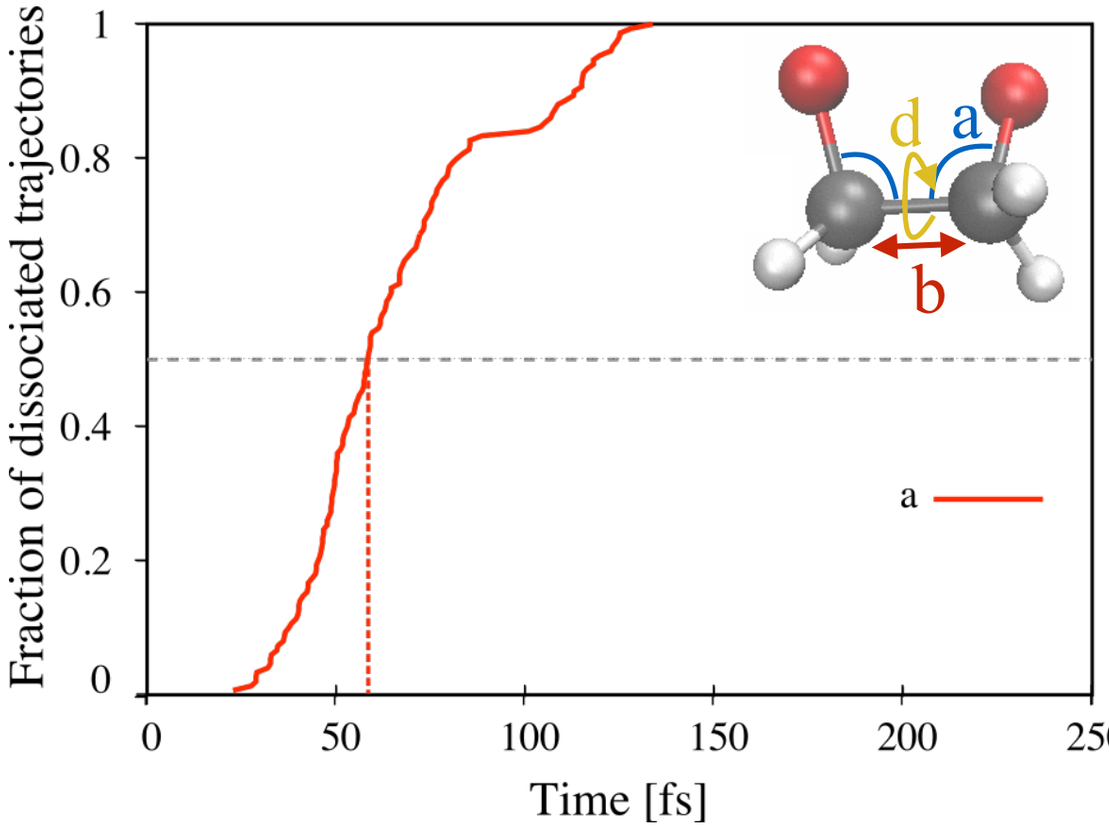
… related to empirical rules known today as:

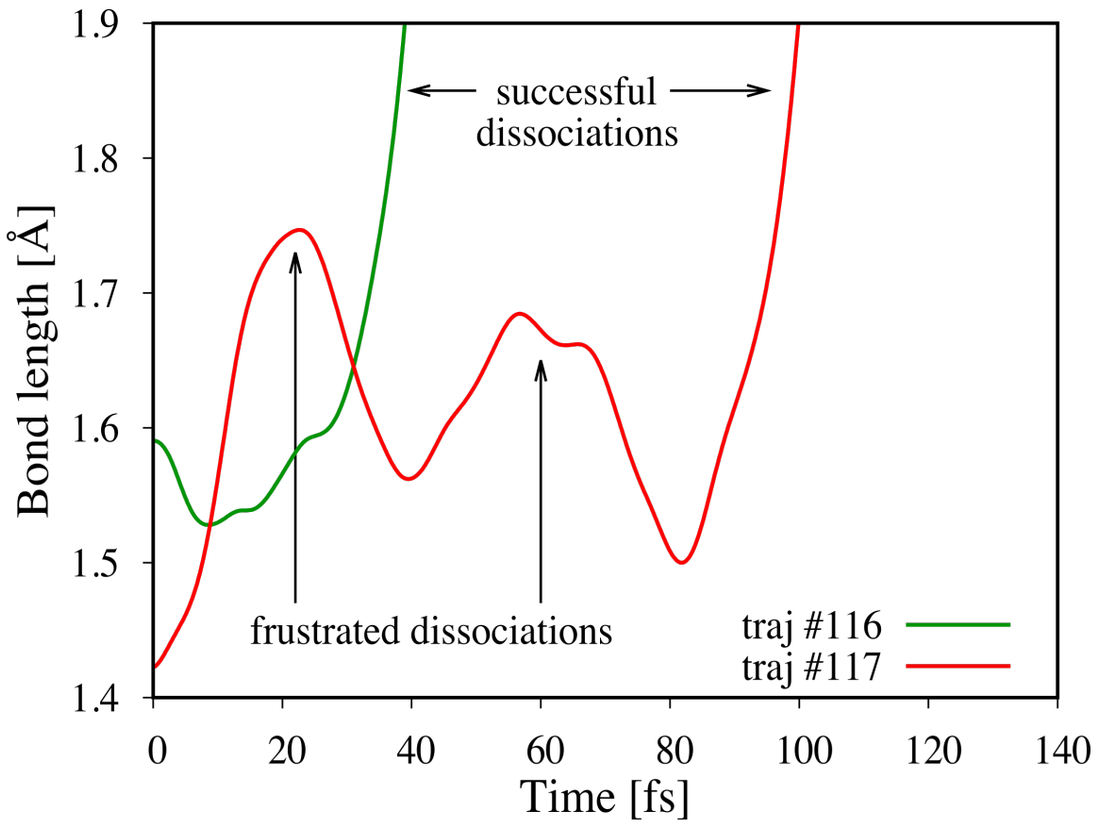
- octet rule
- relation between bond order and bond length
- orbital hybridisation / valence shell electron pair repulsion (VSEPR) model

This is chemistry !

Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *Chem. Science*, **10**, 2298-2307 (2019)

“Frustrated” dissociations





➜ Dissociation time scale

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➜ Geometrical conditions necessary

O-C-C-O dihedral > 55°

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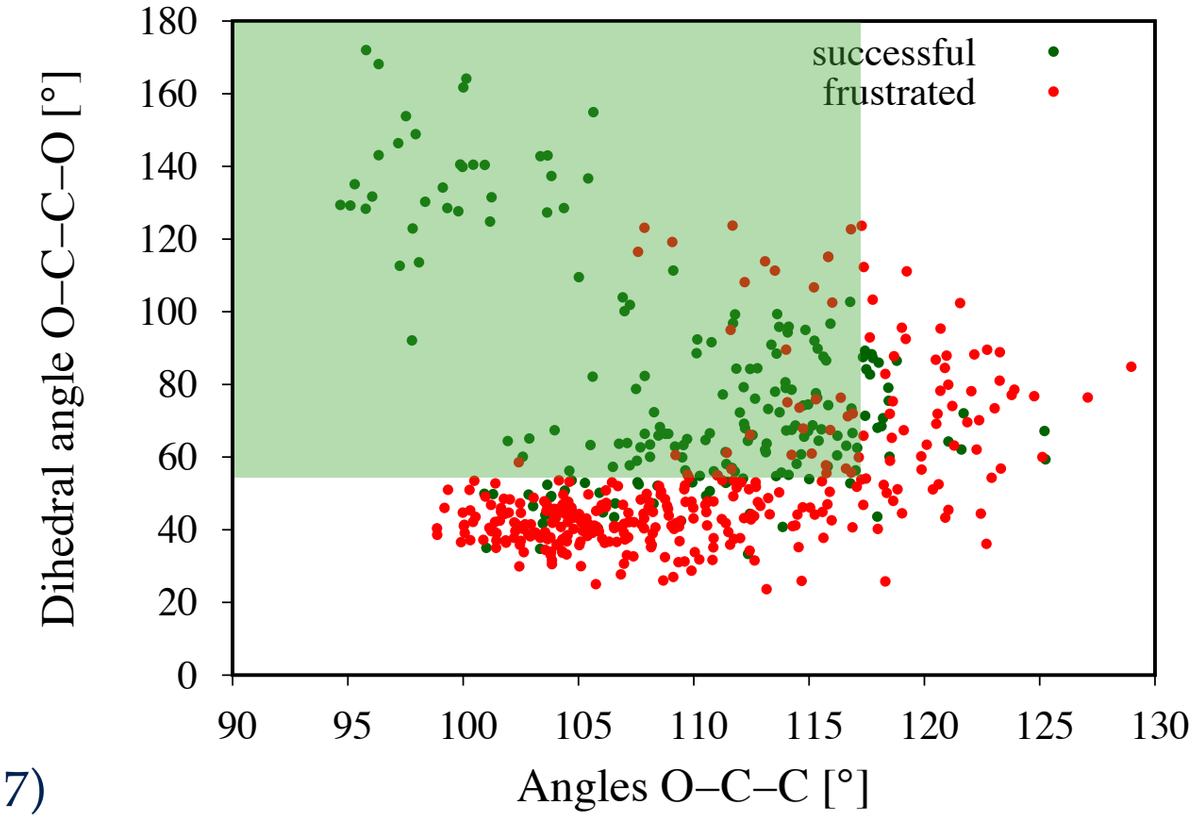
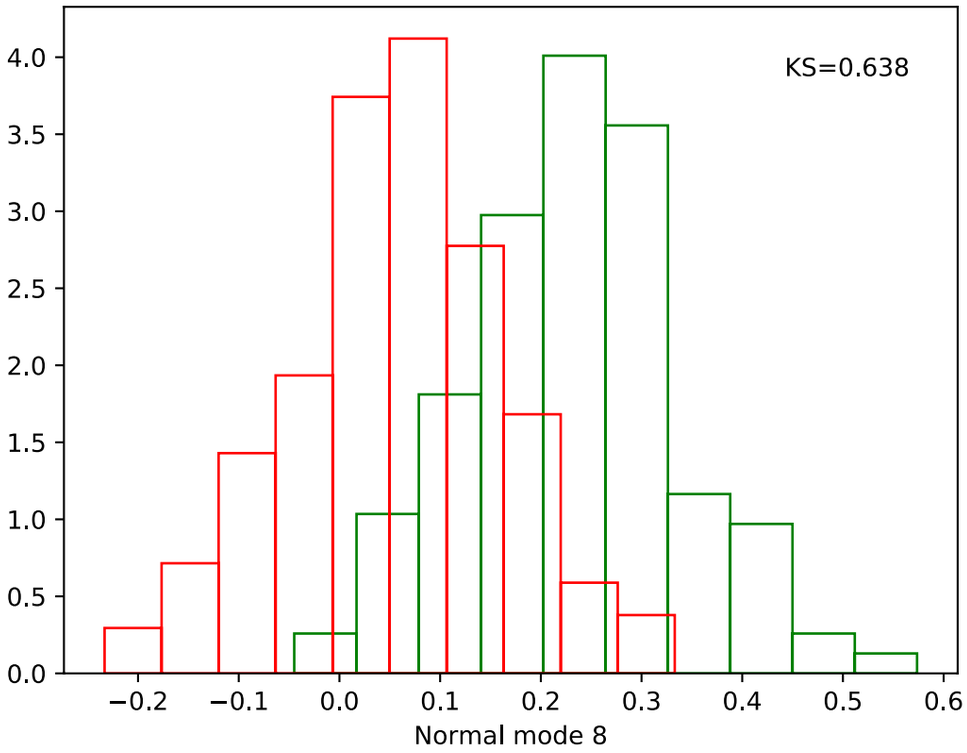
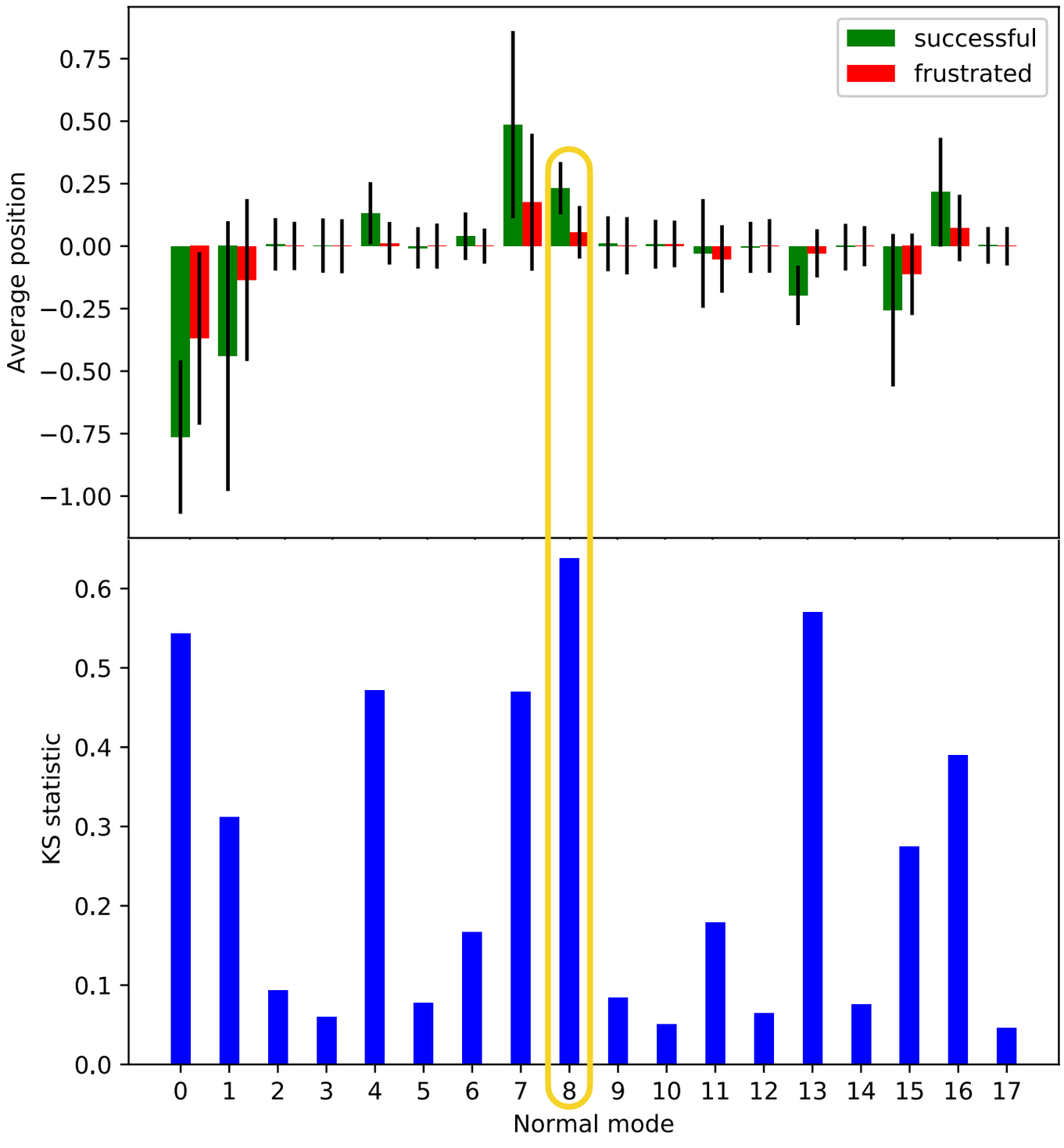


Vacher et al, *J. Chem. Theory Comput.*, **13** (6), 2448-2457 (2017)

“Frustrated” dissociations - revisited





KS = Kolmogorov–Smirnov

Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *J. Phys. Conf. Series*, **1412**, 042003 (2020)

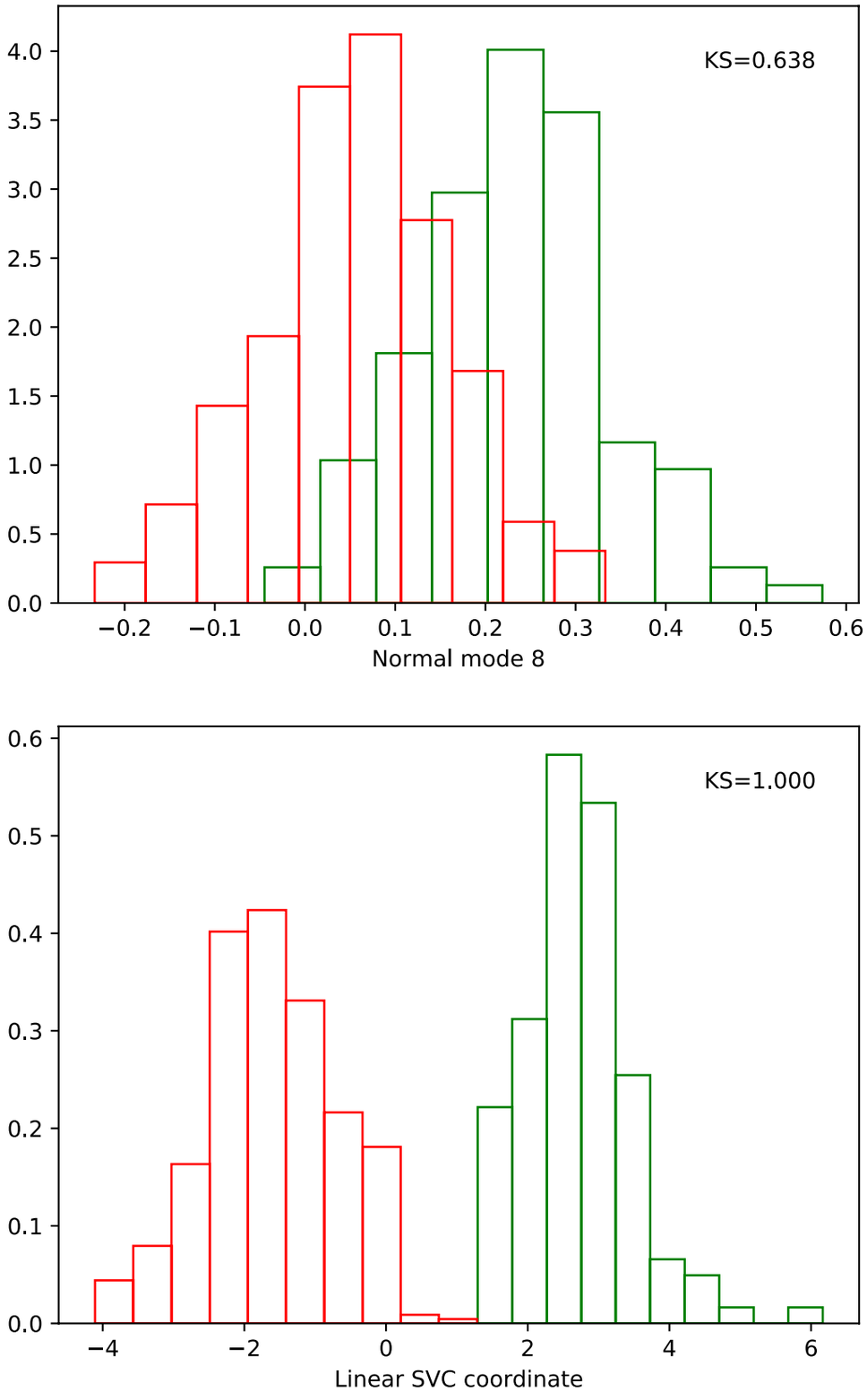
“Frustrated” dissociations - revisited

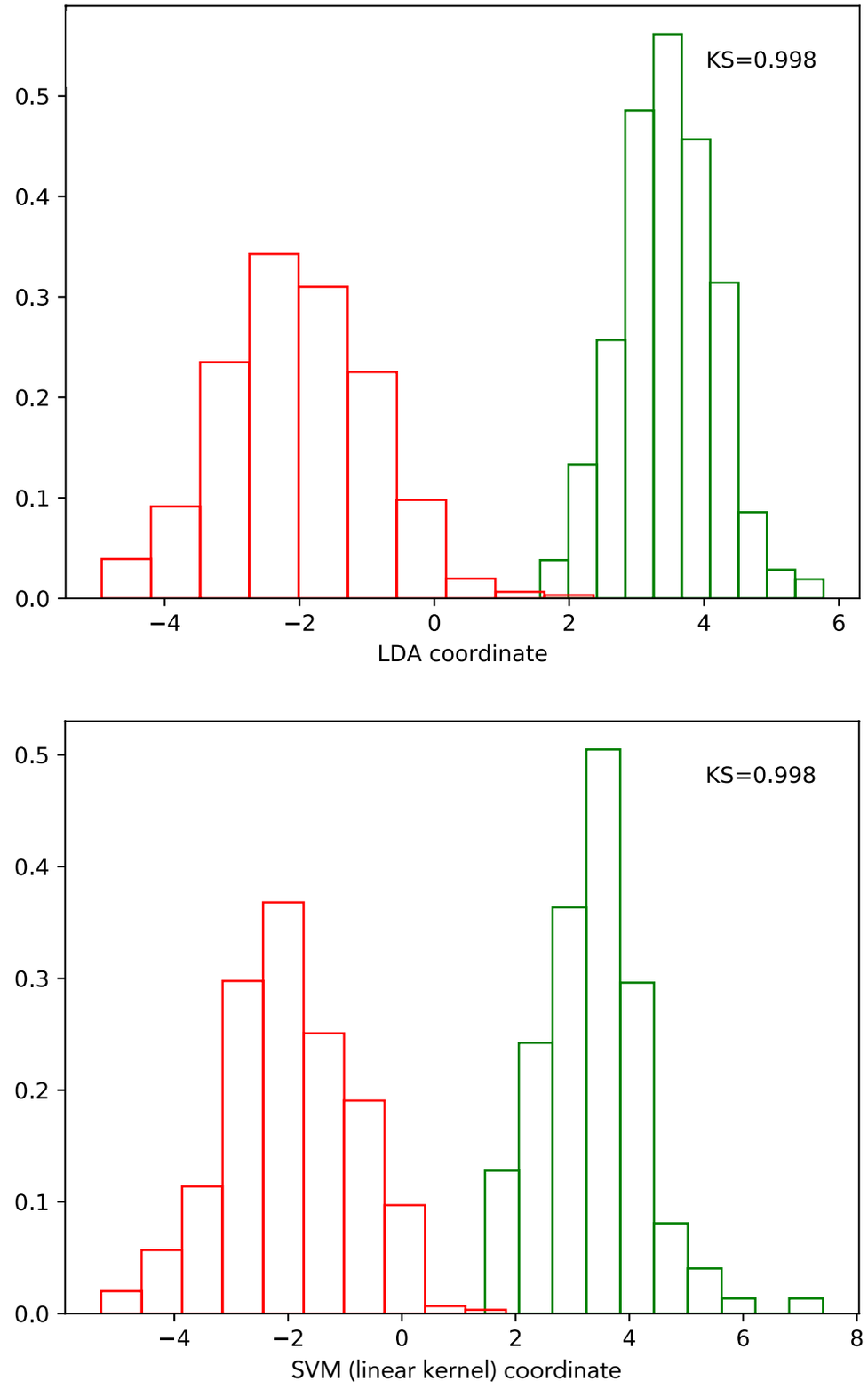
Three different standard machine learning classifiers:

- linear discriminant analysis (LDA)
- linear support vector classification (SVC)
- support vector machines (SVM) with a linear kernel

Häse, Fdez Galván, Aspuru-Guzik, Lindh and Vacher, *J. Phys. Conf. Series,* **1412**, 042003 (2020)

“Frustrated” dissociations - revisited





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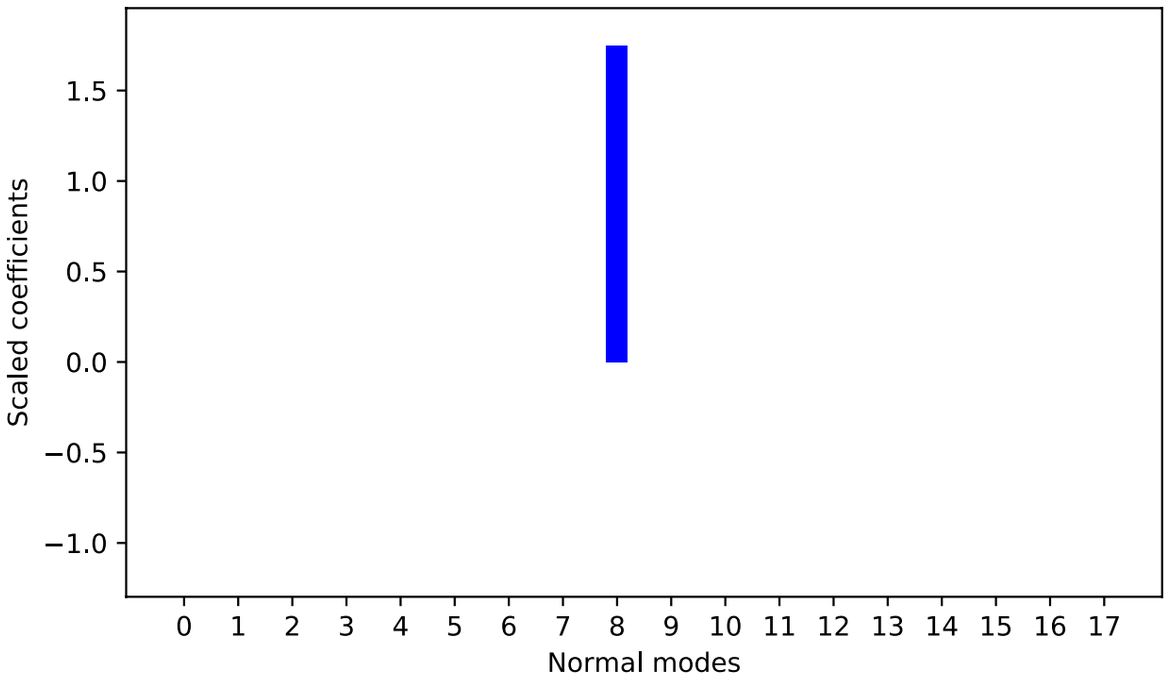
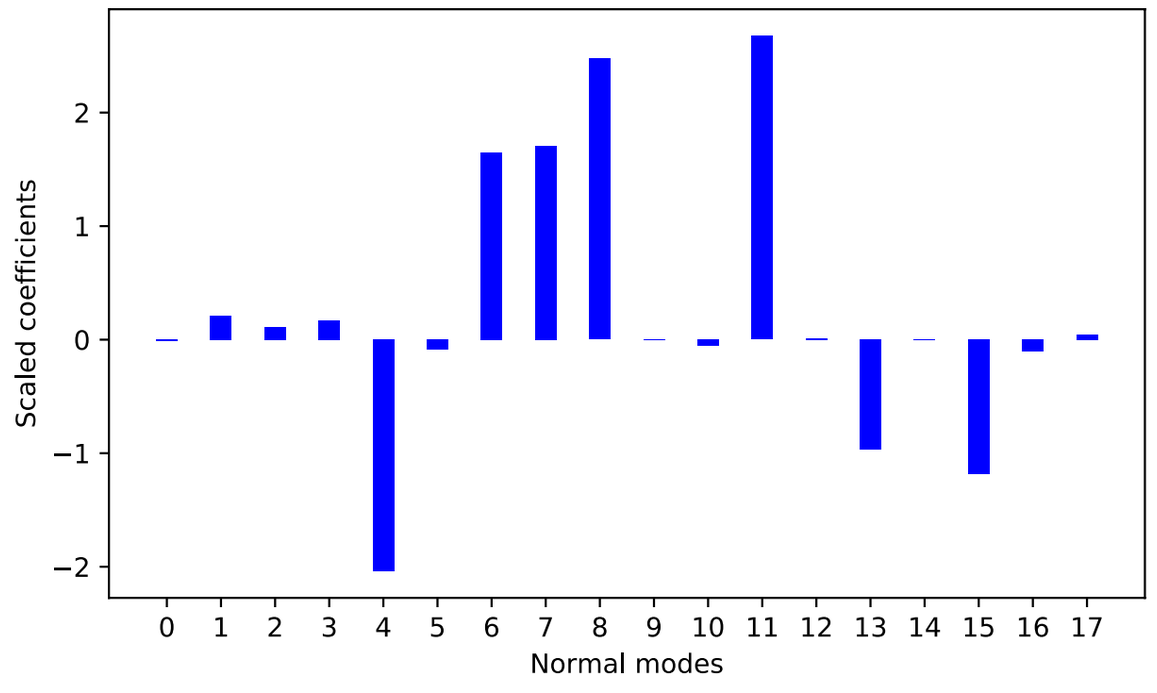
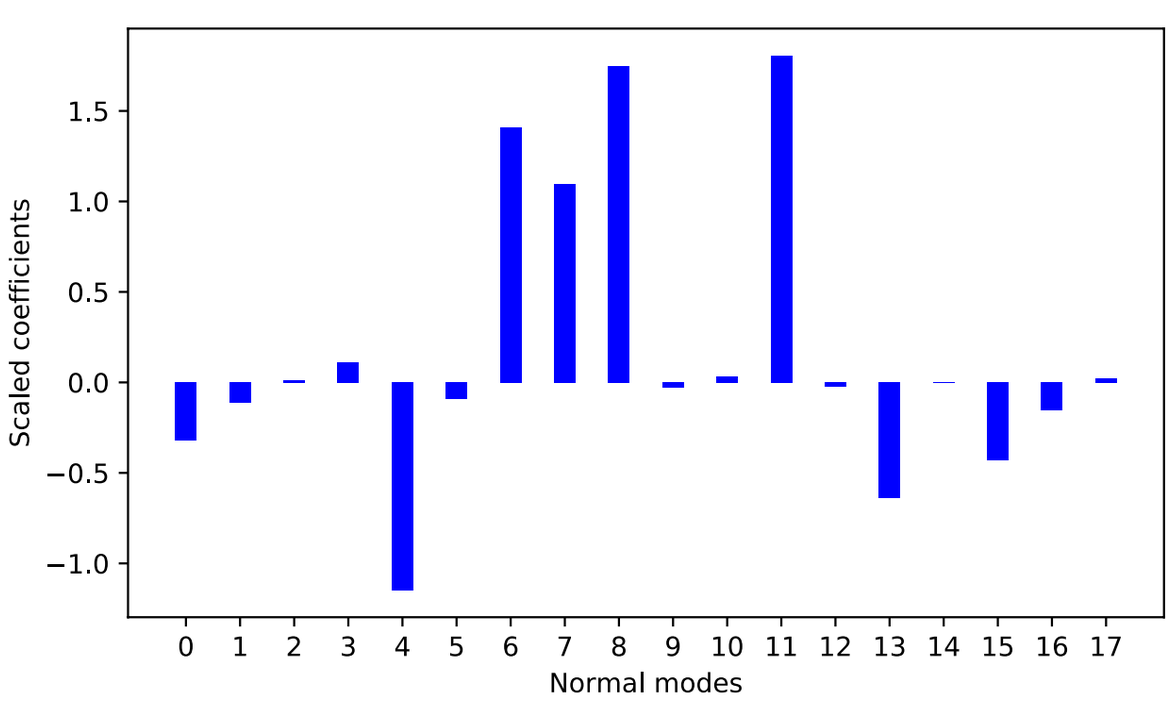
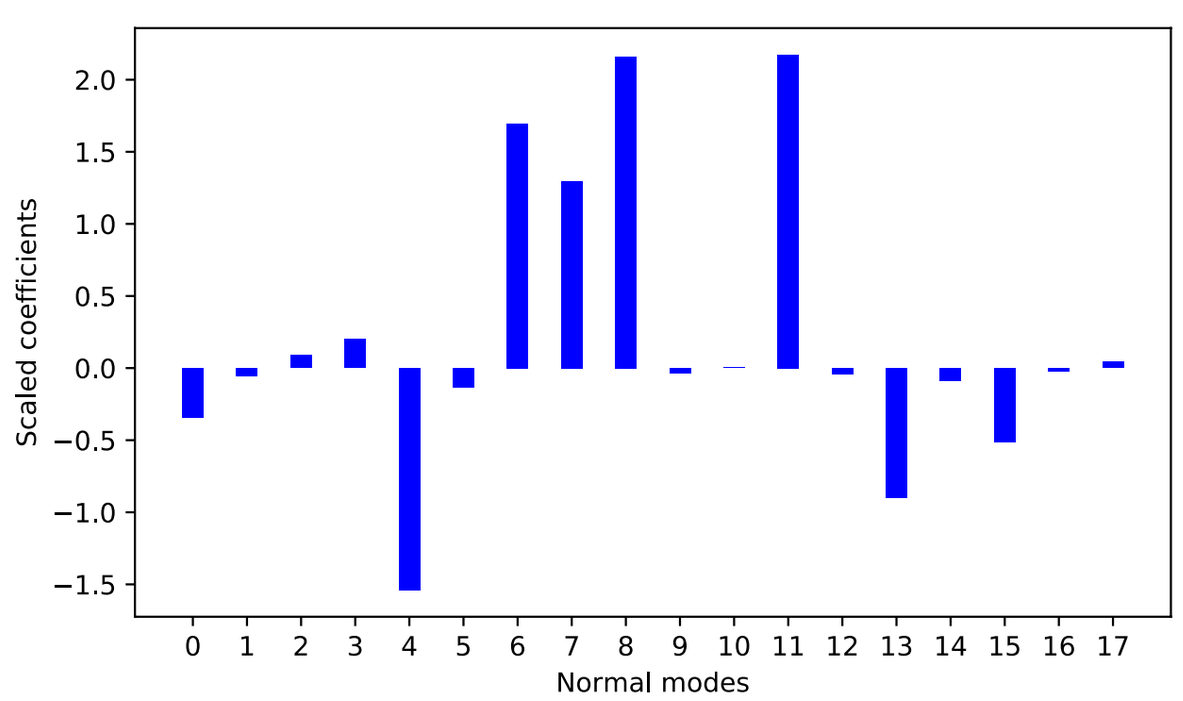
“Frustrated” dissociations - revisited









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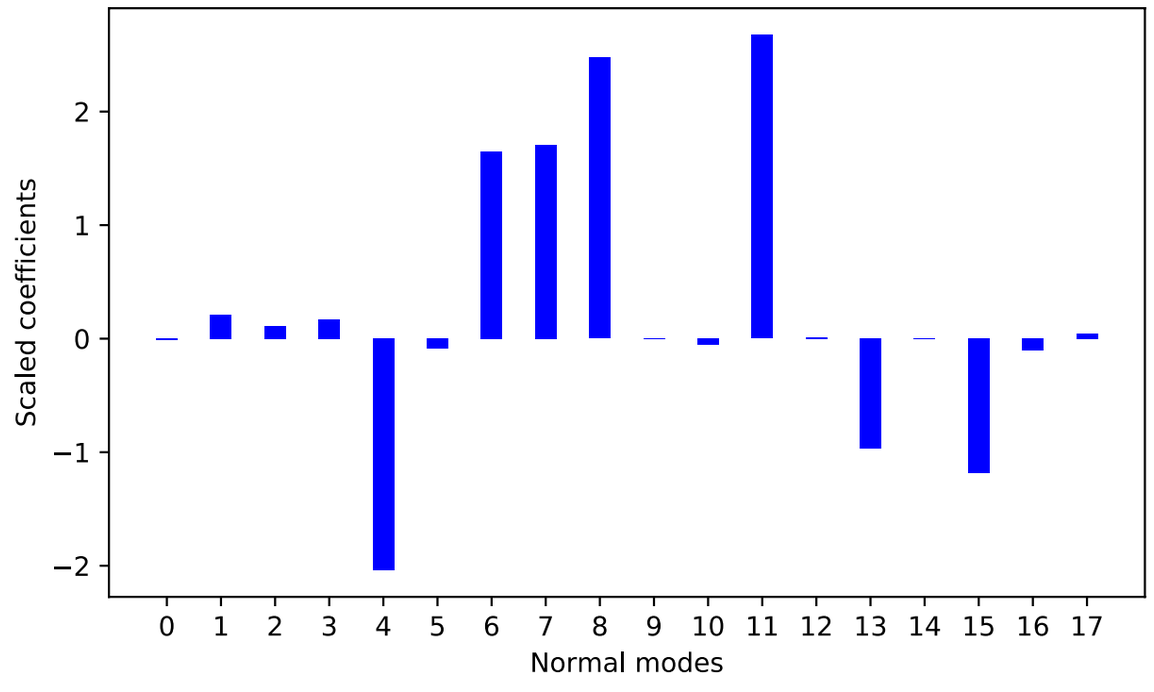
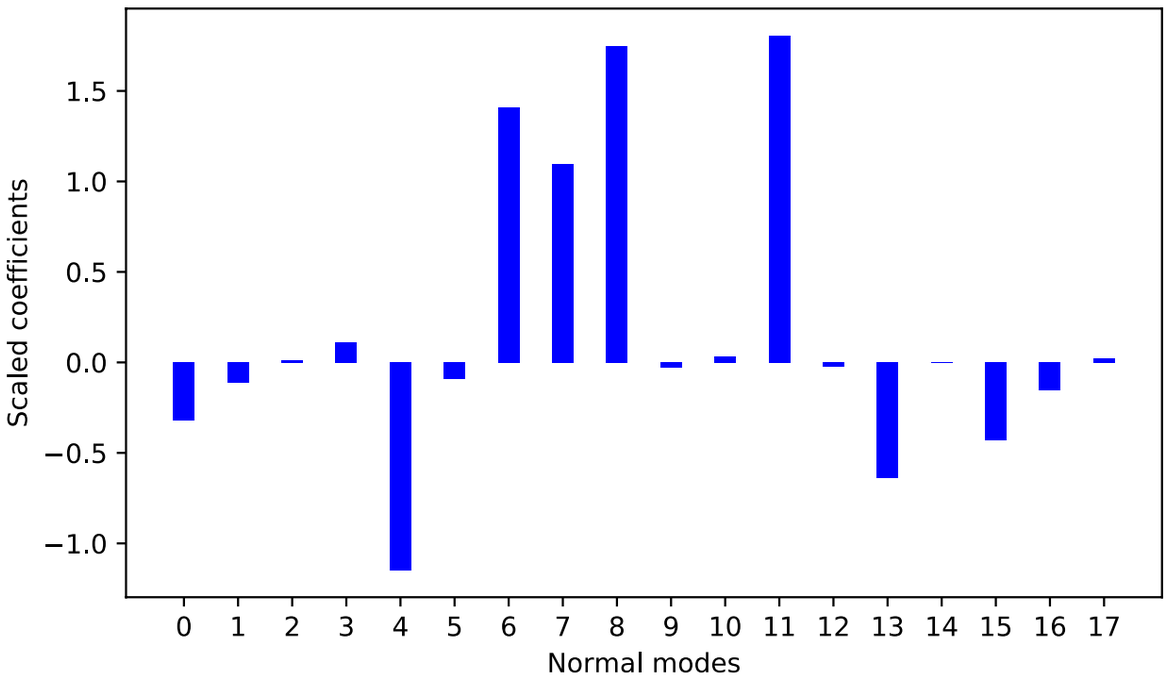
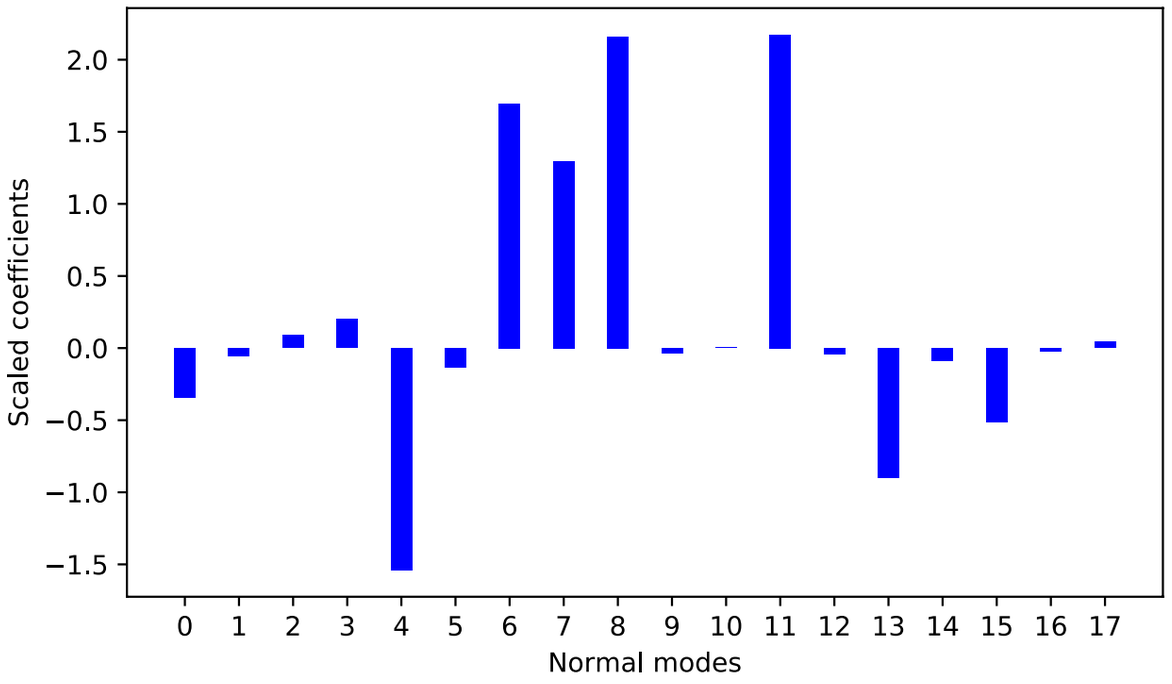
“Frustrated” dissociations - revisited

➜ In-phase planarisation motion of the two formaldehyde moieties







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Take home messages

- Ab initio molecular dynamics simulations are necessary to provide details into the mechanisms and yields of photochemical reactions.

- Machine learning algorithms are able to predict accurately a specific outcome quantity of AIMD simulations. In order to make accurate predictions, the models evidence empirical rules that are, today, part of the common chemical knowledge.

- Machine learning techniques are also helpful to analyse and further interpret the results produced by the AIMD simulations.

- This paves the way for new conceptual insights in chemistry where machine analysis would provide a source of inspiration for us.

Acknowledgements

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Thank you!

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