XACS
Xiamen Atomistic Computing Suite
XACScloud.com

MLatom.com



ML for spectroscopy

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dr-dral.com

1. The Understanding of H_2

2. Computing of F_2

3. Resonance in C_6H_6

4. Computing of O_2

5. Computing of post-VBSCF methods

6. Computing of diabatic states with VB theory

7. Menshutkin Reaction $NH_3 + CH_3Cl \rightarrow [NH_3CH_3]^+ + Cl^-$

8. Charge-shift bonding in propellane

MACHINE LEARNING

1. Machine learning basics

2. ML for PES

3. Universal machine learning models

4. Spectroscopy

4.1. (Ro)vibrational spectra

4.2. UV/vis absorption spectra

4.3. Emission spectra

4.4. Two-photon absorption spectra

4. Spectroscopy

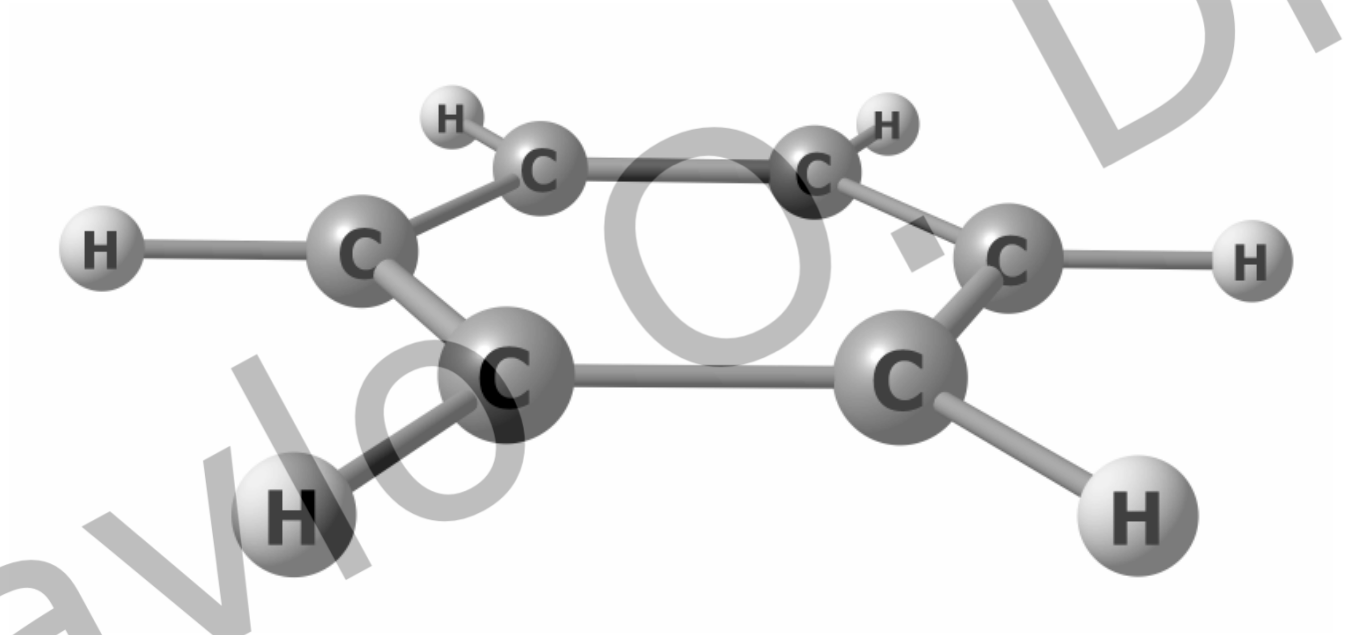
Spectroscopy simulations are very important for bridging the theoretical understanding of molecular structure and reactivity with the experimental measurements. QM spectroscopy is typically quite expensive and here ML can help both as a surrogate, faster replacement of QM methods or be used to directly predict spectra. Also, there is long-standing interest in using ML for interpreting spectra and, most importantly, to obtain structures from spectra, but this is much more difficult and less mature. Here we can't cover all types of spectroscopy but show on several common types how they can be simulated and what we should pay attention to when setting up the simulations.

- (Ro)vibrational spectra
  - IR from frequency calculations
  - IR and power spectra from MD
- UV/vis absorption spectra
  - UV/vis spectra from single-point calculations
  - Nuclear-ensemble approach
- Emission spectra
- Two-photon absorption spectra

4.1. (Ro)vibrational spectra

Rotational-vibrational (rovibrational), i.e., infrared (IR) and Raman, spectroscopy provides a direct way to probe the molecular structure. Hence, such spectra simulations have big practical importance. Their accurate simulations with QM methods are, however, rather expensive. ML is used to significantly accelerate such simulations

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IR spectra from static calculations

$$H_{ij} = \frac{\partial^2 E}{\partial R_i \partial R_j}$$

Normal modes are eigenvectors (obtained by diagonalization)

Eigenvalues are force constants k

Frequencies are derived from force constants as:

$$\nu \sim \sqrt{\frac{k}{\mu_r}}$$

$$I_n = \sum_{t=1}^{3} \left(\frac{\partial p_t}{\partial Q_n} \right)^2 ,$$

$$I_n = \sum_{t=1}^{3} \left(\sum_x \frac{\partial p_t}{\partial q_x} \frac{\partial q_x}{\partial Q_n} \right)^2 ,$$

Note

Currently, MLatom supports dipole derivatives calculations via MNDO, Gaussian and Pyscf.

Calculations done within the rigid-rotor harmonic oscillator approximation.

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Example ML-spectra-1.

Calculate IR spectrum of ethanol molecule with MLatom using static approach.

1. Analyze output file. How many normal modes does the molecule have?
2. Does the spectrum agrees well with experiment?

```
ir
uaiqm_wb97x631gp@cc
xyzfile=opt.xyz
```

where the `opt.xyz` is the ethanol optimized by `uaiqm_wb97x631gp@cc` as:

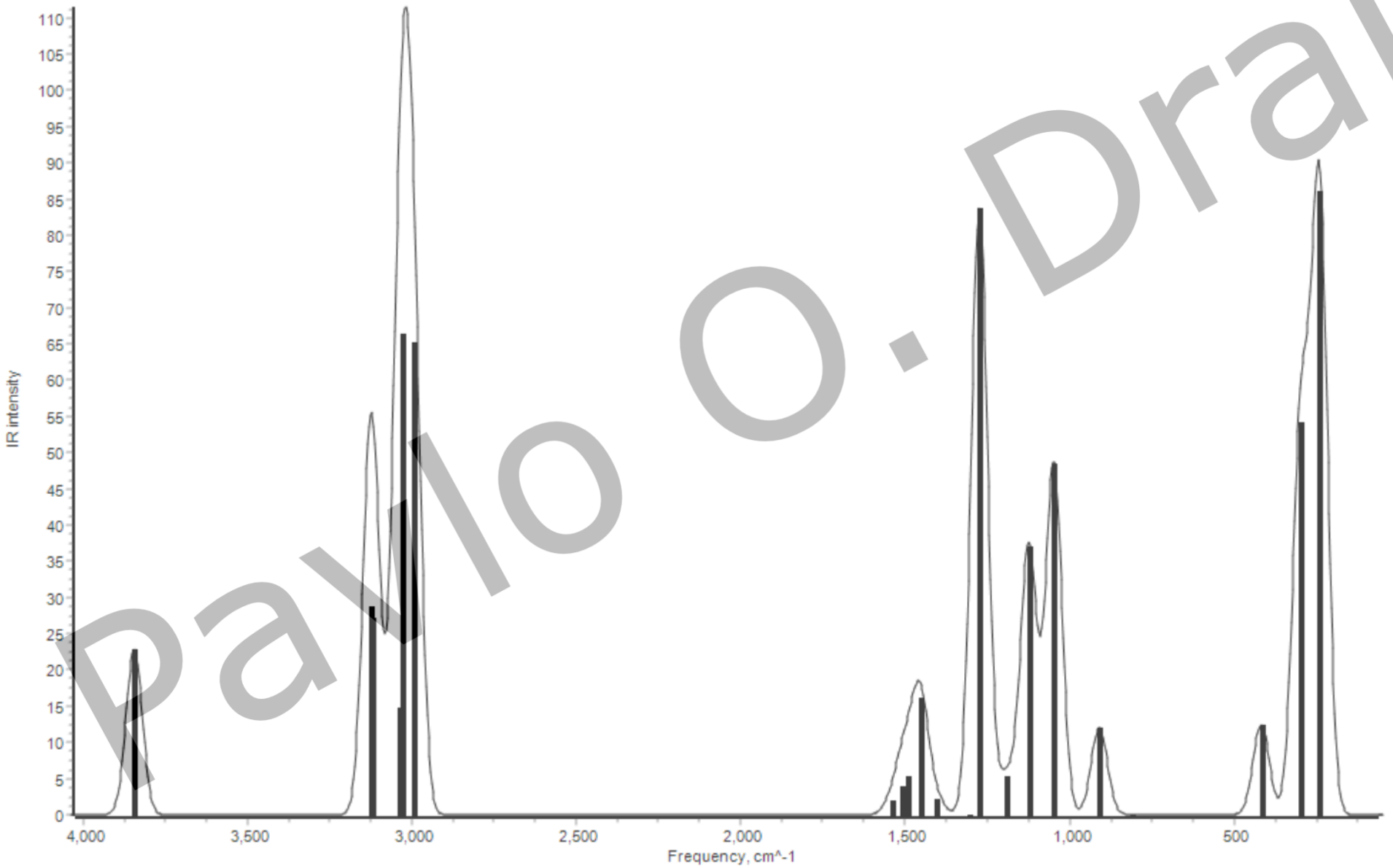
```
9

C          1.2141461144075        -0.2237522351565         0.0000009660331
H          1.2725810451795        -0.8571489137580        -0.8839209229985
H          1.2725955610560        -0.8571196861893         0.8839423252131
H          2.0641316742868         0.4586264688194        -0.0000176008701
C         -0.0820255073362         0.5506969112630        -0.0000006009490
H         -0.1394716683438         1.1910011723471         0.8856982674255
H         -0.1394676608187         1.1909953759585        -0.8857037188954
O         -1.1431943032587        -0.3953014469416        -0.0000014007109
H         -1.9775381677168         0.0743891017157         0.0000106653069
```

Warning

Always remember to optimize your molecule at the same level!

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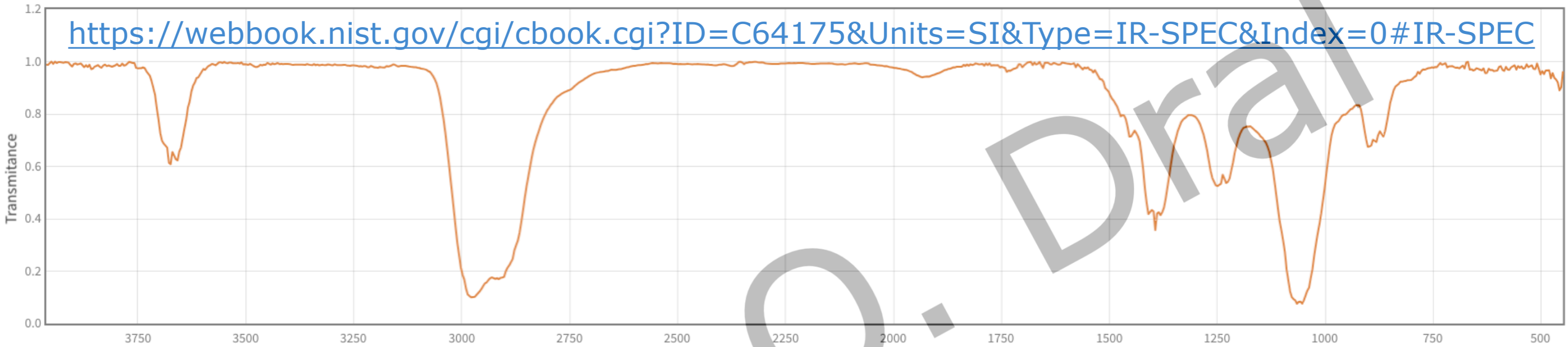

IR intensity
110
105
100
95
90
85
80
75
70
65
60
55
50
45
40
35
30
25
20
15
10
5
0
4,000
3,500
3,000
2,500
2,000
1,500
1,000
500
Frequency, cm^-1

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Ethanol

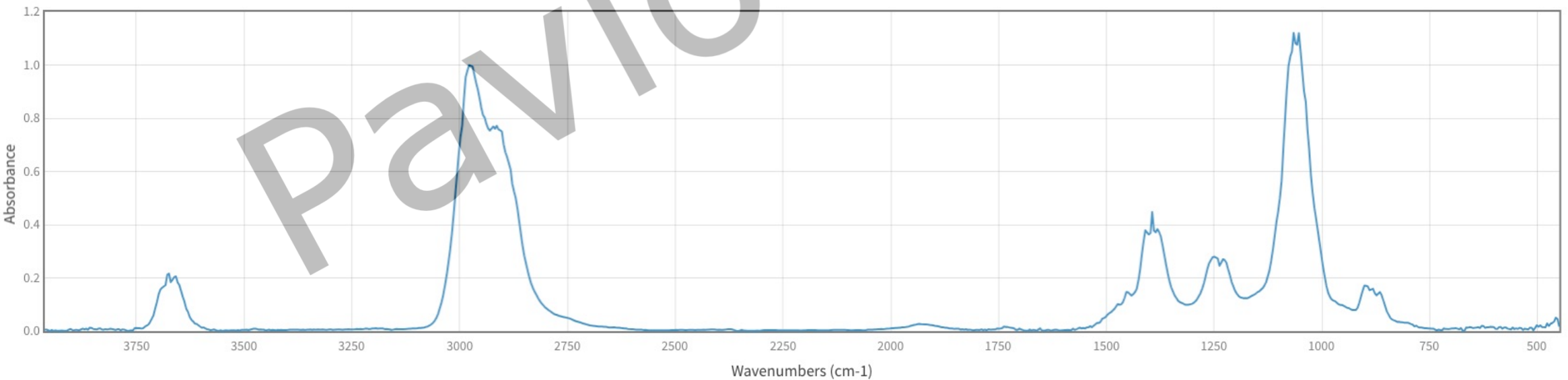
Infrared Spectrum

https://webbook.nist.gov/cgi/cbook.cgi?ID=C64175&Units=SI&Type=IR-SPEC&Index=0#IR-SPEC



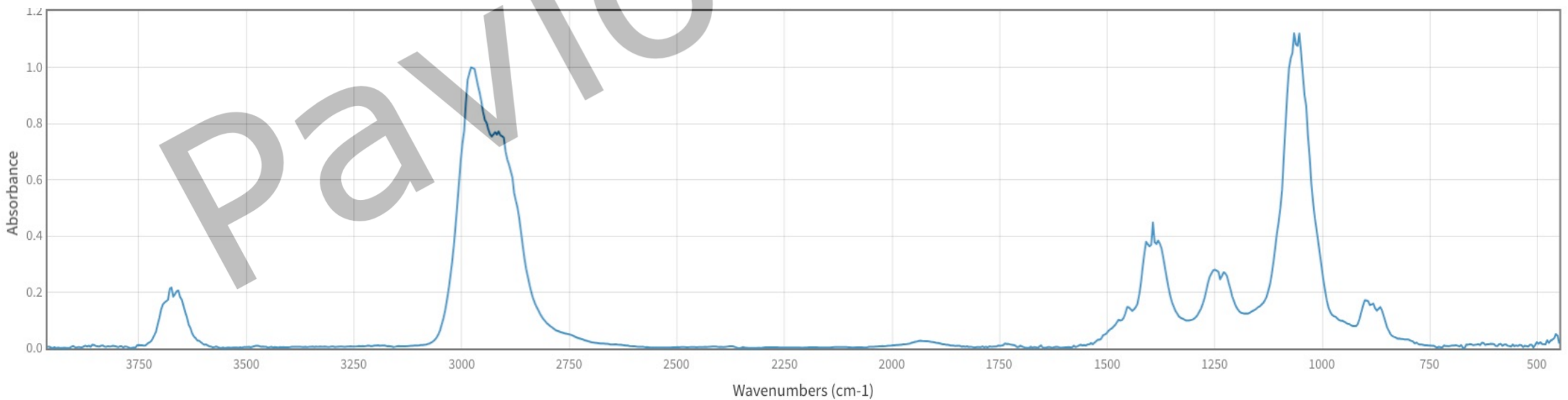
Ethanol

Infrared Spectrum



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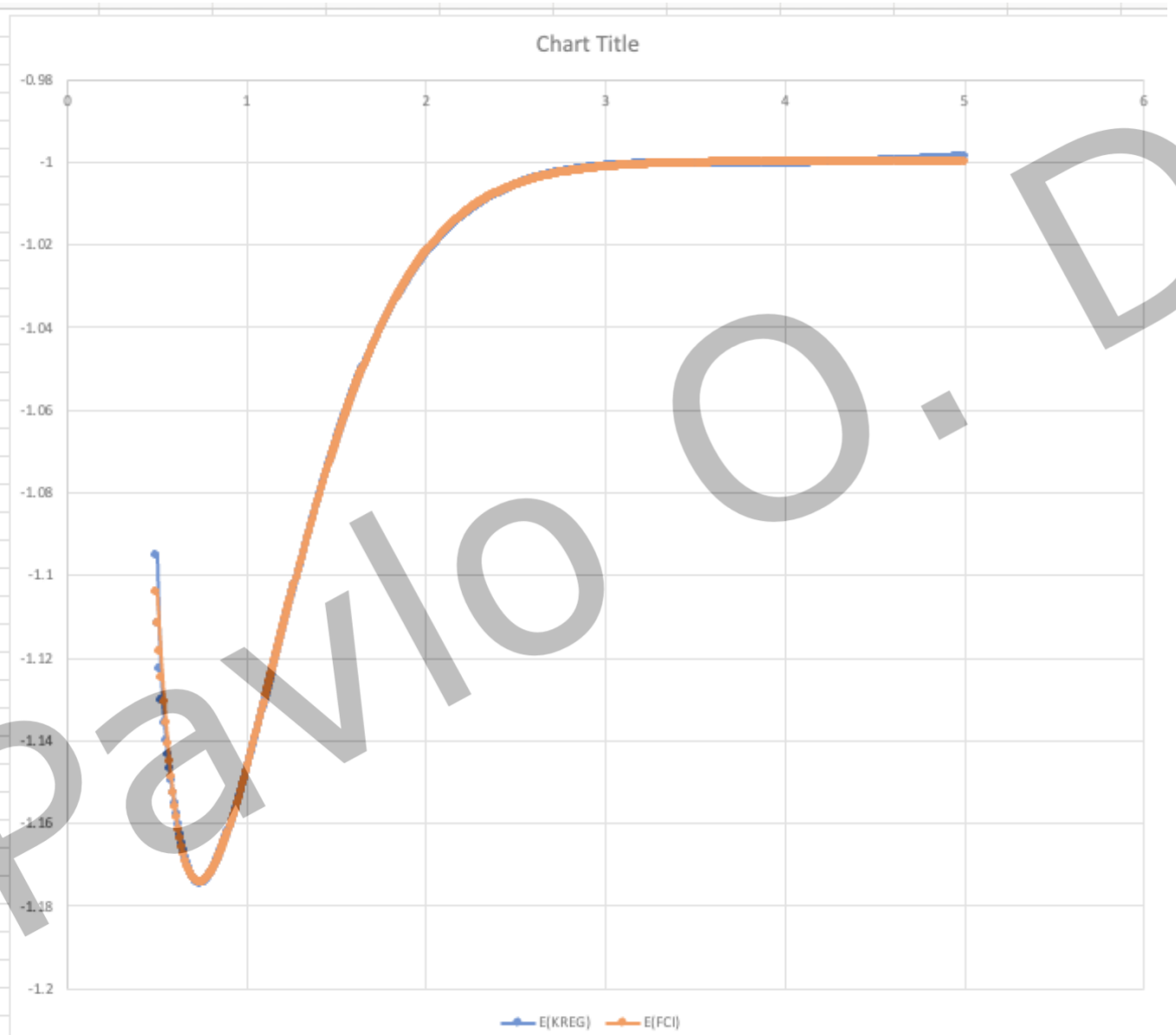




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Actual spectra have anharmonicity effects

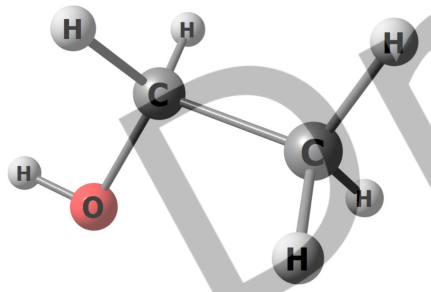
R	E(KREG)	E(FCI)
0.5	-1.0952055	-1.1041324
0.51	-1.1116607	-1.1116602
0.52	-1.1226388	-1.1185659
0.53	-1.1302139	-1.1248928
0.54	-1.1357334	-1.1306807
0.55	-1.1400459	-1.1359664
0.56	-1.1436606	-1.1407837
0.57	-1.1468859	-1.145164
0.58	-1.1498826	-1.1491365
0.59	-1.1527206	-1.1527279
0.6	-1.1554383	-1.1559632
0.61	-1.1580248	-1.1588655
0.62	-1.1604676	-1.1614566
0.63	-1.1627427	-1.1637562
0.64	-1.1648372	-1.1657832
0.65	-1.1667376	-1.1675548
0.66	-1.1684246	-1.1690873
0.67	-1.1698978	-1.1703957
0.68	-1.1711619	-1.1714941
0.69	-1.1722143	-1.1723958
0.7	-1.1730593	-1.1731131
0.71	-1.1737061	-1.1736575
0.72	-1.1741656	-1.1740399
0.73	-1.1744488	-1.1742703
0.74	-1.1745676	-1.1743585
0.75	-1.1745317	-1.1743132
0.76	-1.1743587	-1.1741428
0.77	-1.1740494	-1.1738553
0.78	-1.1736193	-1.173458
0.79	-1.1730809	-1.1729579
0.8	-1.1724471	-1.1723616
0.81	-1.1717182	-1.1716753
0.82	-1.1708958	-1.1709047
0.83	-1.1700126	-1.1700554
0.84	-1.1690501	-1.1691325
0.85	-1.1680262	-1.1681409



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Infrared spectra from MD

Long MD trajectory is needed!



Power spectrum:

$$P(\omega) \propto \int \langle \boldsymbol{v}(\tau)\boldsymbol{v}(t+\tau)\rangle_\tau e^{-i\omega t}\mathrm{d}t$$

Autocorrelation function of velocities

IR intensities:

$$P(\omega) \propto \omega \tanh\left(\frac{\hbar\omega}{2k_B T}\right)\int \langle \boldsymbol{\mu}(\tau)\boldsymbol{\mu}(t+\tau)\rangle_\tau e^{-i\omega t}\mathrm{d}t$$

Fast Fourier transform of the autocorrelation function of dipole moments

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L. Zhang & Y. Hou, F. Ge, P. O. Dral, *Phys. Chem. Chem. Phys.* **2023**, *25*, 23467

Example ML-spectra-2.

Calculate IR spectrum of ethanol molecule with MLatom from an MD trajectory which was calculated at AIQM1 level for 10 ps with 0.5 time step.

1. How does the spectrum compares to the spectrum from static calculations?
2. Compared to experiment, what are the major qualitative differences?

Input file:

```
MD2vibr                      # 1. requests vibrational spectra from MD trajectory
trajH5MDin=ethanol_traj.h5   # 2. file with MD trajectory in h5md format
dt=0.5                       # 3. time step of trajectory; unit: fs
start_time=0                 # 4. use trajectory starting from 0 fs
end_time=10000               # 5. use trajectory ending at 10000 fs
autocorrelationDepth=1024    # 6. autocorrelation depth; unit: fs
zeropadding=1024             # 7. zero padding length; unit: fs
output=ir                    # 8. output infrared spectrum
```

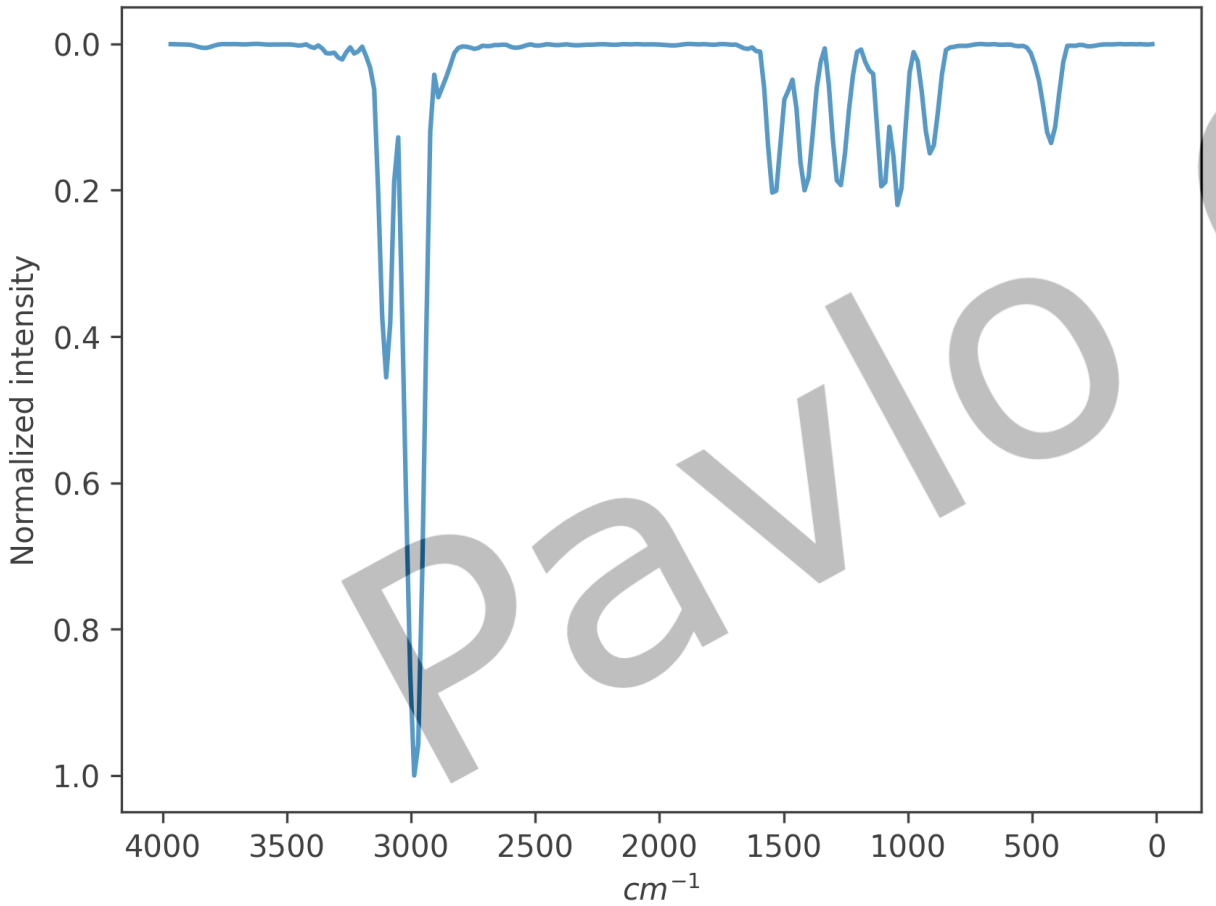
Auxiliary files:

- ethanol_traj.h5 - trajectory in the h5md format, required for calculations.
- ethanol_traj.xyz - if you want to check how the trajectory looks like (not necessary for calculations)

After calculations finish, you will find `ir.png` with the plot of the infrared spectrum.

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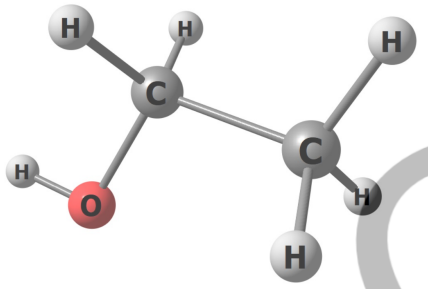
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zeropadding=1024                 # 7. zero padding length; unit: fs
output=ir                        # 8. output infrared spectrum
```



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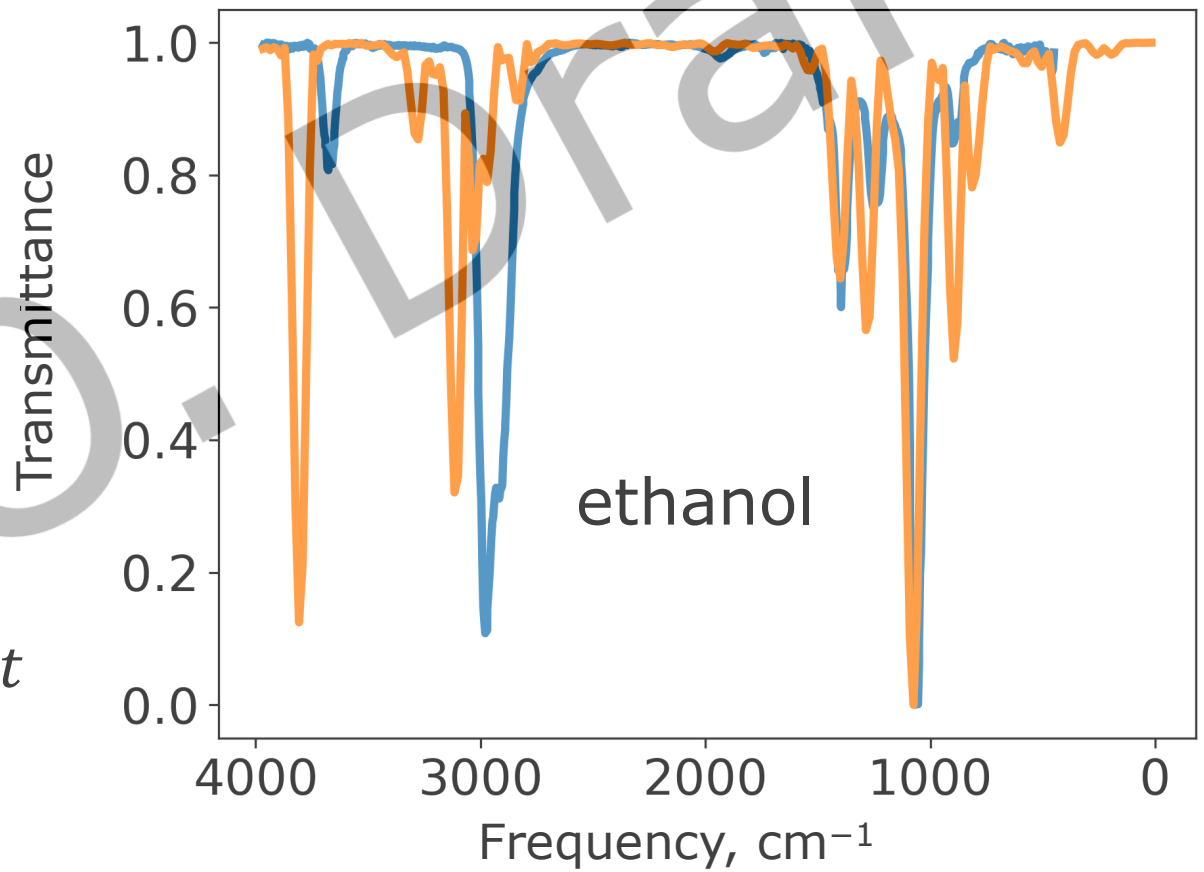
Infrared spectra from MD

Long MD trajectory is needed!



IR intensities:

$$P(\omega) \propto \omega \tanh\left(\frac{\hbar\omega}{2k_B T}\right) \int \langle \boldsymbol{\mu}(\tau)\boldsymbol{\mu}(t+\tau)\rangle_\tau e^{-i\omega t} \mathrm{d}t$$

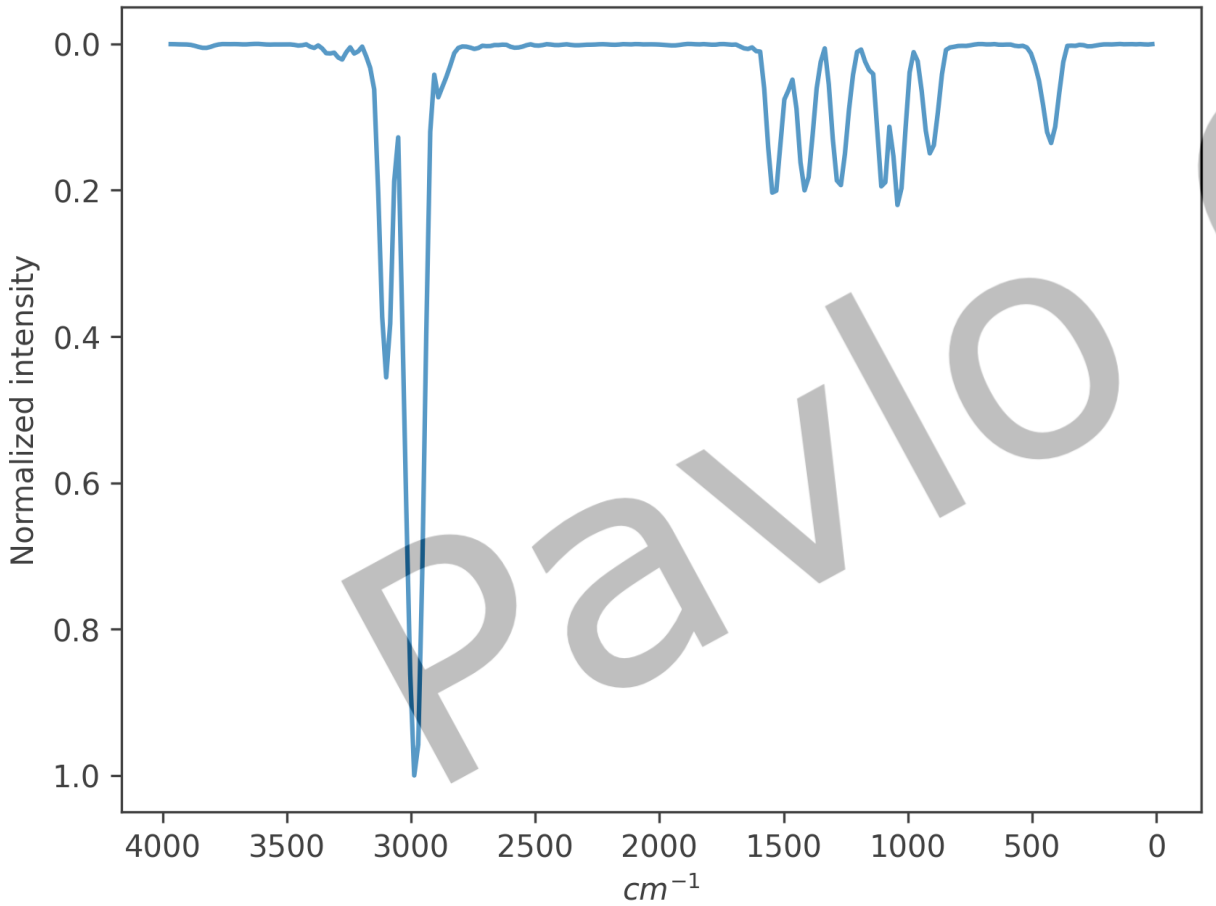


Experiment (NIST)
AIQM1 MD
200 ps trajectory at 300 K

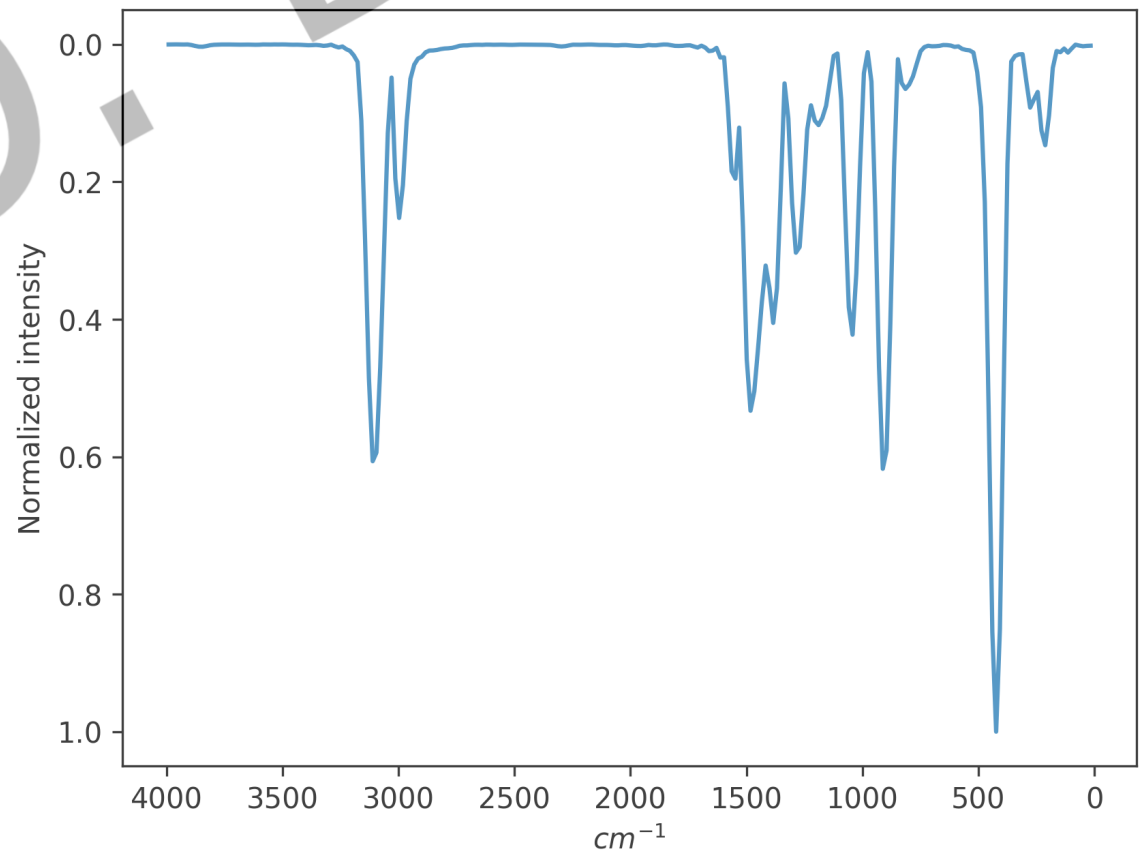
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L. Zhang & Y. Hou, F. Ge, P. O. Dral, *Phys. Chem. Chem. Phys.* **2023**, *25*, 23467

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end_time=10000                   # 5. use trajectory ending at 10000 fs
autocorrelationDepth=1024        # 6. autocorrelation depth; unit: fs
zeropadding=1024                 # 7. zero padding length; unit: fs
output=ir                        # 8. output infrared spectrum
```

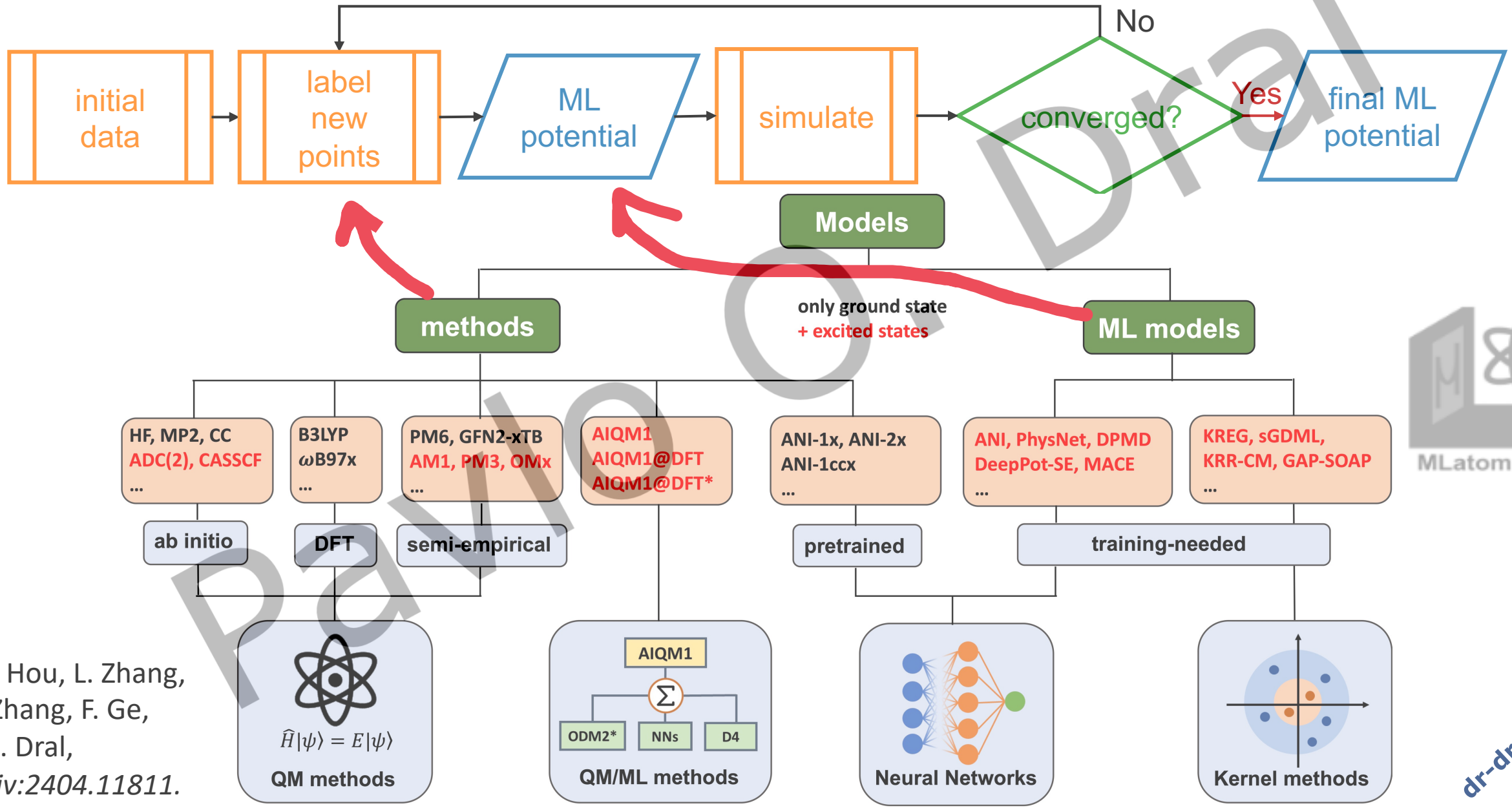



```
MD2vibr                          # 1. requests vibrational spectra from MD trajectory
trajH5MDin=ethanol_traj.h5       # 2. file with MD trajectory in h5md format
dt=0.5                           # 3. time step of trajectory; unit: fs
start_time=0                     # 4. use trajectory starting from 0 fs
end_time=10000                   # 5. use trajectory ending at 10000 fs
autocorrelationDepth=1024        # 6. autocorrelation depth; unit: fs
zeropadding=1024                 # 7. zero padding length; unit: fs
output=ps                        # 8. output power spectrum
```



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Physics-informed active learning



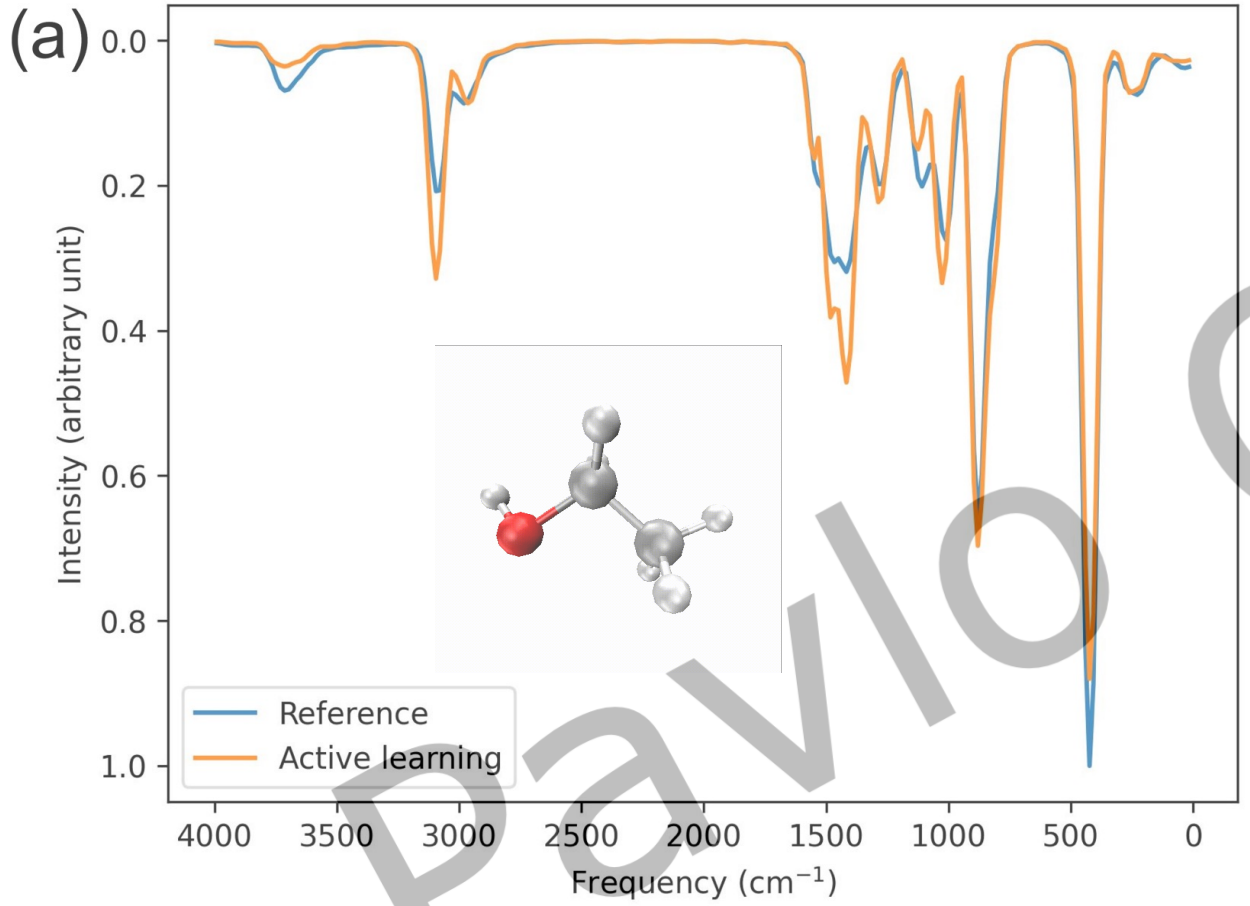
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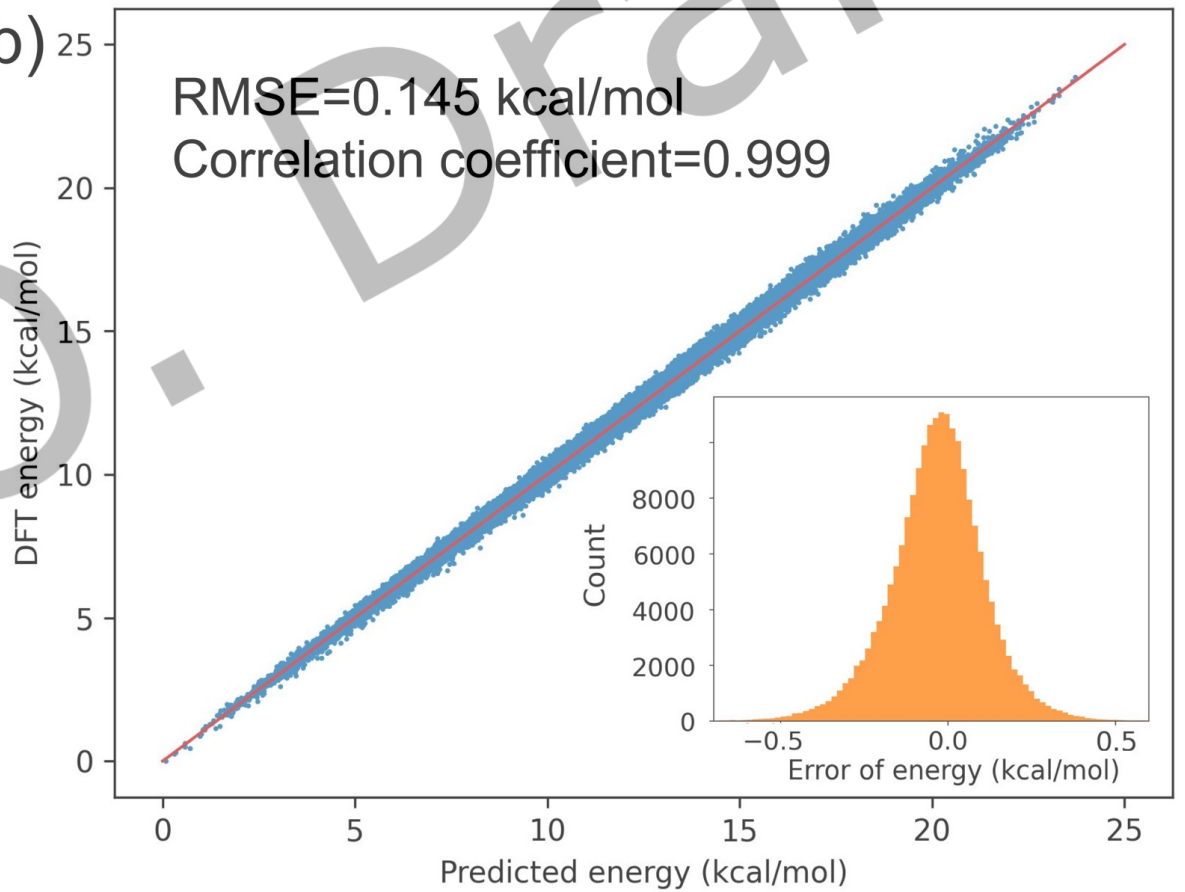
MLatom.com

Y.-F. Hou, L. Zhang, Q. Zhang, F. Ge, P. O. Dral, *arXiv:2404.11811.*

Accurate vibrational spectra

< 1000 training points with ANI potential



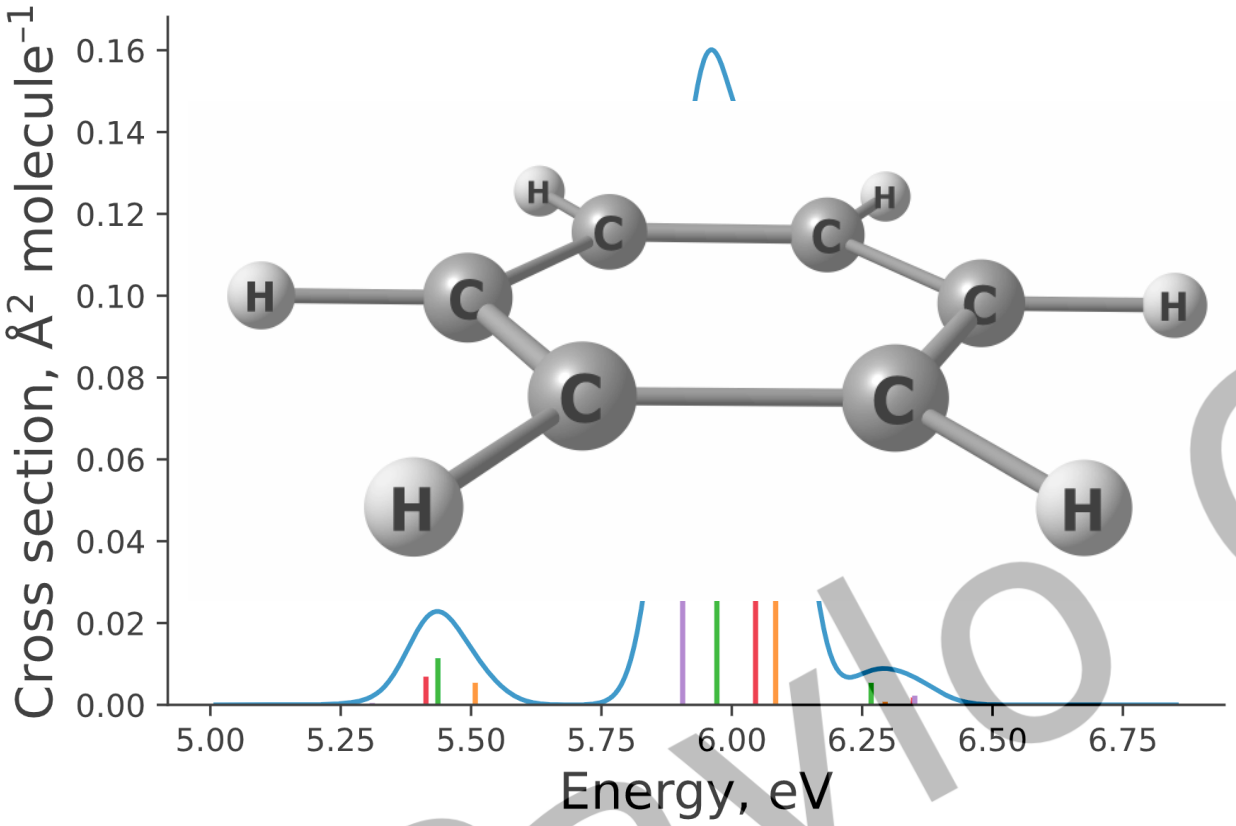


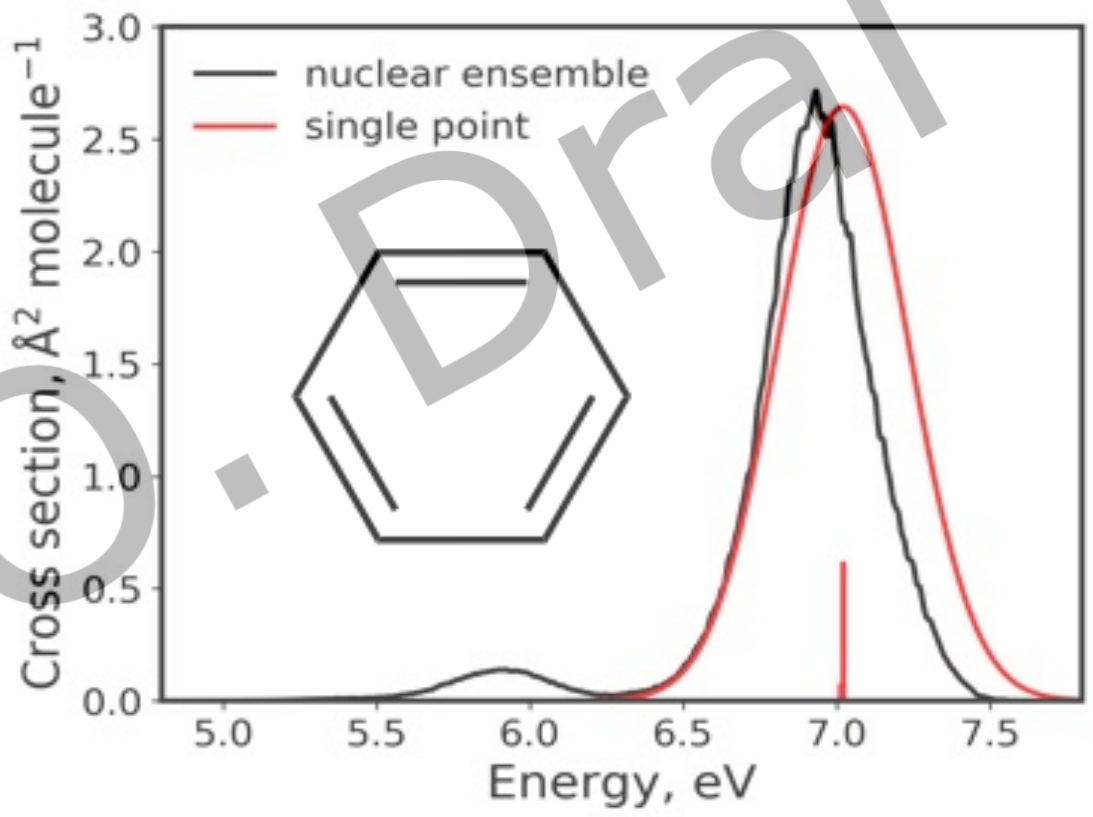
Ethanol spectra

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Y.-F. Hou, L. Zhang, Q. Zhang, F. Ge, P. O. Dral. *arXiv:2404.11811*.

Machine learning UV/vis absorption spectra





Nuclear Ensemble Approach (NEA) calculates cross section by averaging over multiple normalized broadening functions at different conformations (marked by different colors).

Compared with the single point convolution, NEA correctly predicts absorption intensity for forbidden transitions (example: benzene)

$$\sigma(E) = \frac{\pi e^2 h}{2mc\varepsilon_0 E} \sum_n^{N_{fs}} \frac{1}{N_p} \sum_i^{N_p} \Delta E_{0n}(\mathbf{x}_i) f_{0n}(\mathbf{x}_i) \frac{1}{\sqrt{2\pi(\delta/2)^2}} \exp\left(-\frac{(E-\Delta E_{0n})^2}{2(\delta/2)^2}\right)$$

R. Crespo-Otero, M. Barbatti, *Theor. Chem. Acc.* **2012,** *131*, 1237

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Quantum chemistry vs Machine learning

Conventional programming in quantum chemistry:

→ Code for molecular orbitals

→ Code for excitation energies

→ Code for oscillator strengths

→ …

Machine learning (in principle – adaptations required!):

→ The same code for all above

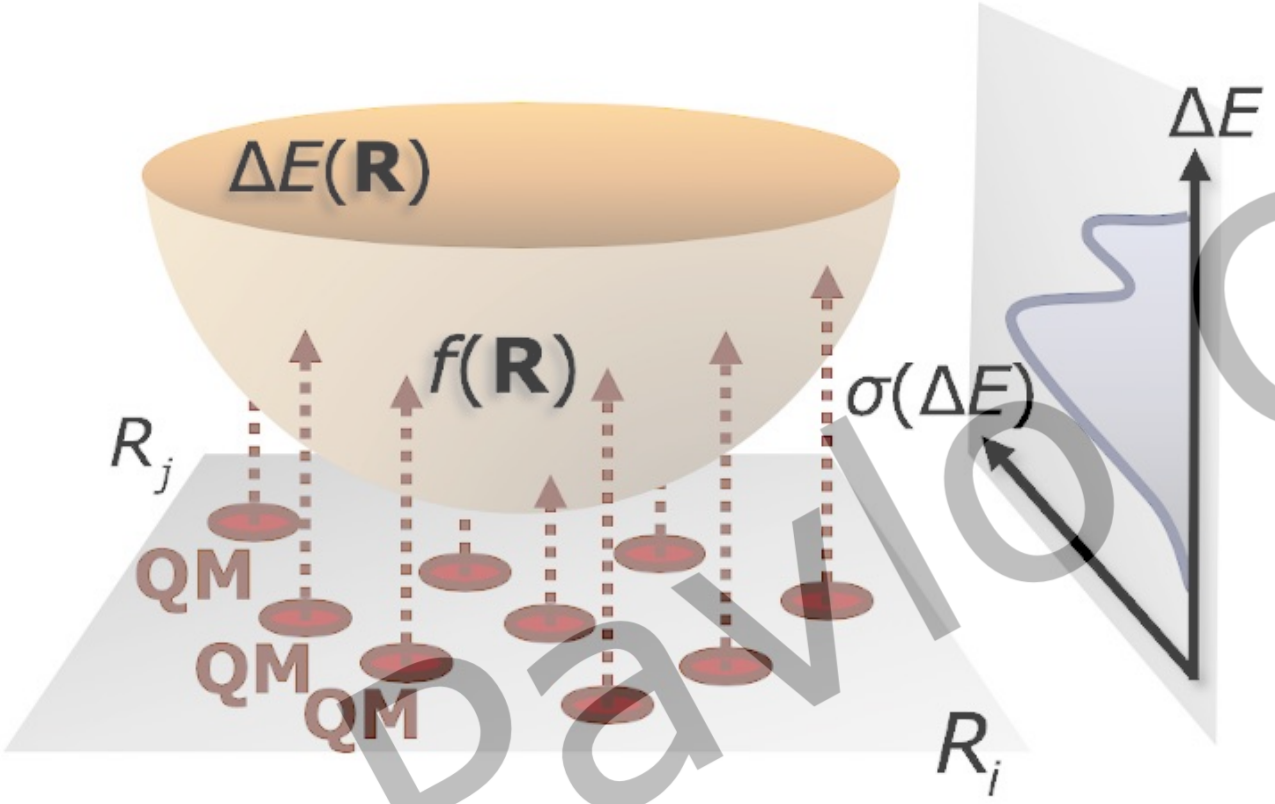
MOs excitation energies Oscillator strength

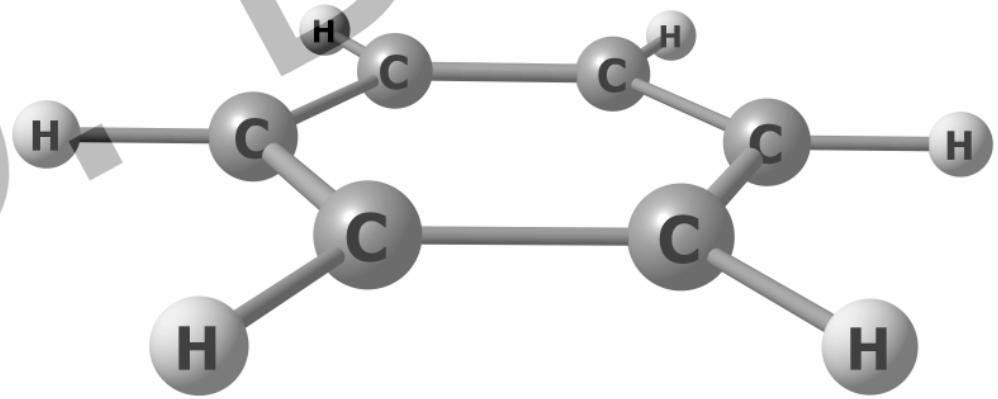
Training data

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ML-nuclear ensemble approach (ML-NEA)

N QM calculations are needed to simulate spectrum



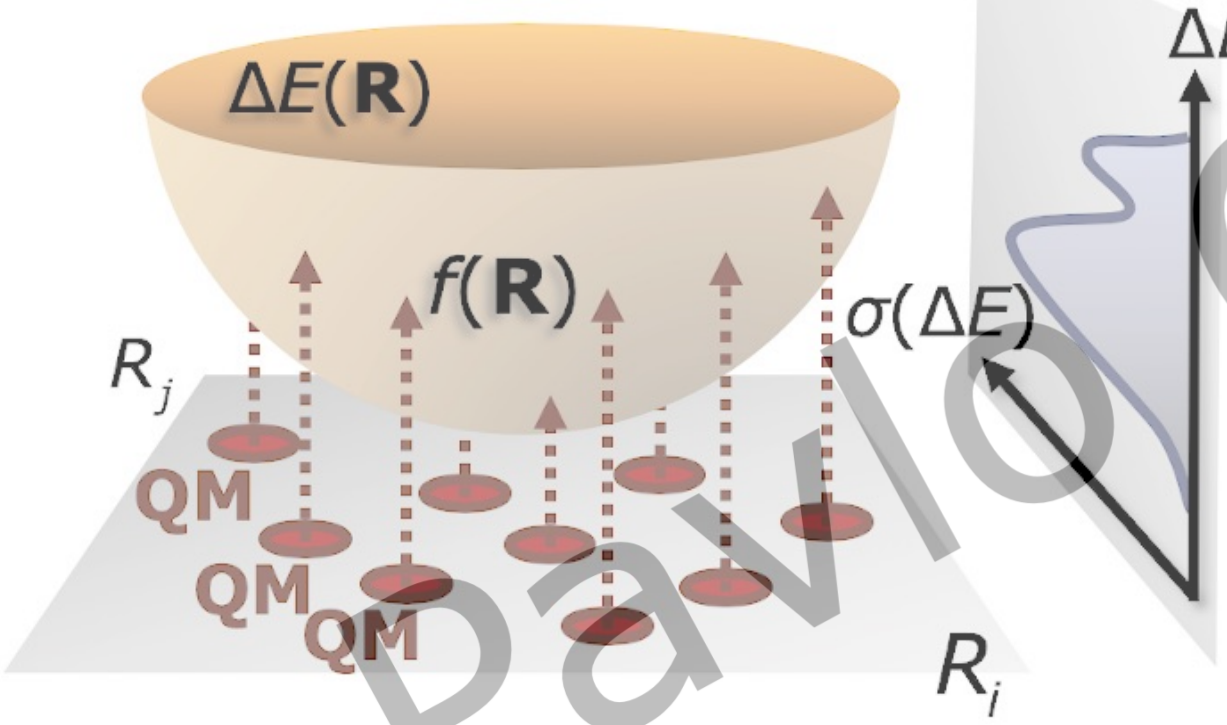


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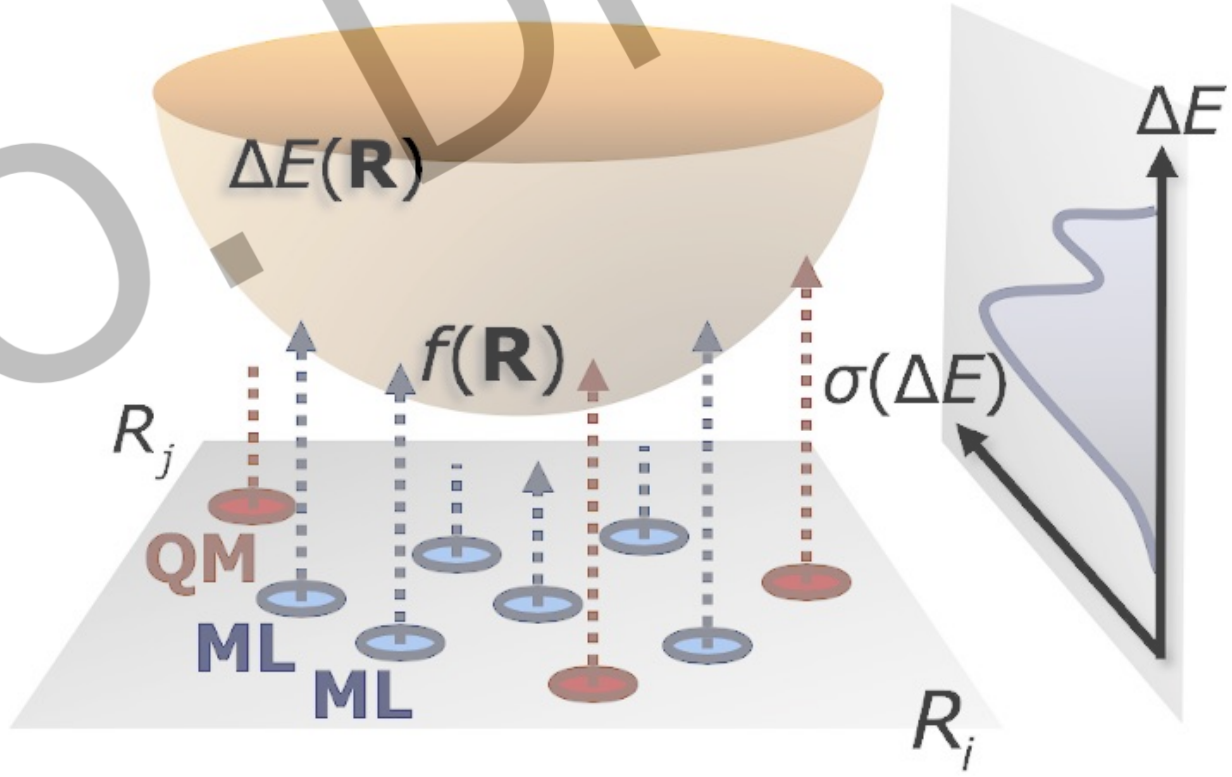
P. O. Dral, M. Barbatti, *Nat. Rev. Chem.* **2021,** *5*, 388

ML-nuclear ensemble approach (ML-NEA)

N QM calculations are needed to simulate spectrum



With ML, only *M* << *N* QM calculations are needed



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P. O. Dral, M. Barbatti, *Nat. Rev. Chem.* **2021,** *5*, 388

ML-nuclear ensemble approach (ML-NEA)

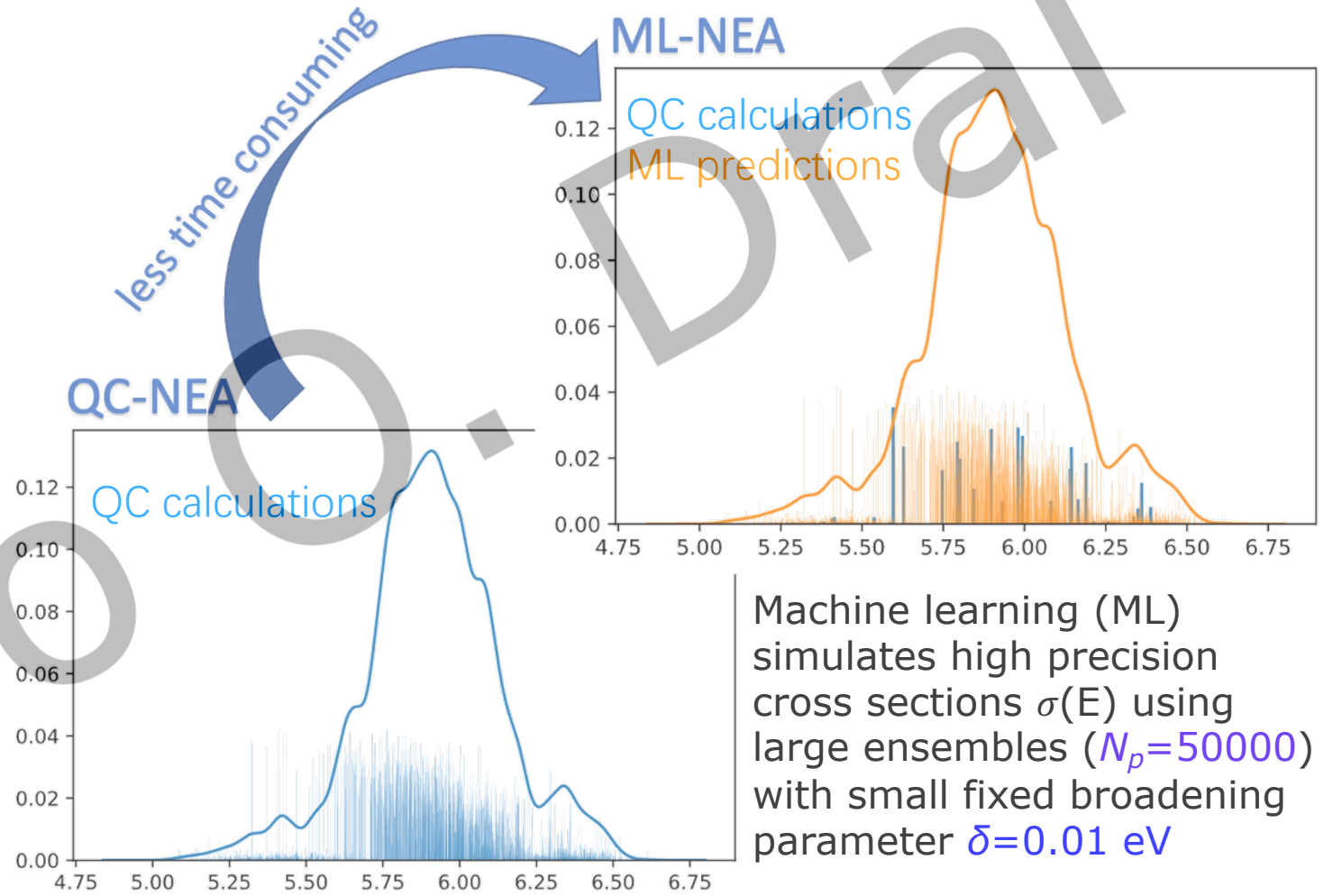
Learned properties:

- excitation energies

$$\Delta E_{0n}(\mathbf{x}_i)$$

- Oscillator strengths

$$f_{0n}(\mathbf{x}_i)$$



Machine learning (ML) simulates high precision cross sections σ(E) using large ensembles (N_p=50000) with small fixed broadening parameter δ=0.01 eV

$$\sigma(E) = \frac{\pi e^2 h}{2mc\varepsilon_0 E} \sum_n^{N_{fs}} \frac{1}{N_p} \sum_i^{N_p} \Delta E_{0n}(\mathbf{x}_i) f_{0n}(\mathbf{x}_i) \frac{1}{\sqrt{2\pi(\delta/2)^2}} \exp\left(-\frac{(E-\Delta E_{0n})^2}{2(\delta/2)^2}\right)$$

B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210

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The KREG model

$$f(\mathbf{x}_i) = \sum_{j=1}^{N_{\text{tr}}} \alpha_j k(\mathbf{x}_i, \mathbf{x}_j)$$

$$k(\mathbf{x}_i, \mathbf{x}_j) = \exp\left(-\frac{1}{2\sigma^2}\sum_{s}^{N_x}(x_{i,s} - x_{j,s})^2\right)$$

the Gaussian kernel function
(σ is the kernel width)

$$x = \begin{pmatrix} \dots & \frac{R^{eq}}{R} & \dots \end{pmatrix}^T$$

RE descriptor

Analytical solution for the regression coefficients α given N_{tr} training points

$$\begin{pmatrix} k(\mathbf{x}_1, \mathbf{x}_1) + \lambda & \cdots & k\left(\mathbf{x}_1, \mathbf{x}_{N_{\text{tr}}}\right) \\ \vdots & \ddots & \vdots \\ k\left(\mathbf{x}_{N_{\text{tr}}}, \mathbf{x}_1\right) & \cdots & k\left(\mathbf{x}_{N_{\text{tr}}}, \mathbf{x}_{N_{\text{tr}}}\right) + \lambda \end{pmatrix} \begin{pmatrix} \alpha_1 \\ \vdots \\ \alpha_{N_{\text{tr}}} \end{pmatrix} = \begin{pmatrix} y_1 \\ \vdots \\ y_{N_{\text{tr}}} \end{pmatrix}$$

λ is the regularization parameter ensuring transferability

$$(\mathbf{K} + \lambda\mathbf{I})\boldsymbol{\alpha} = \mathbf{y}$$

Sub-training set | Validate → for tuning hyperparameters

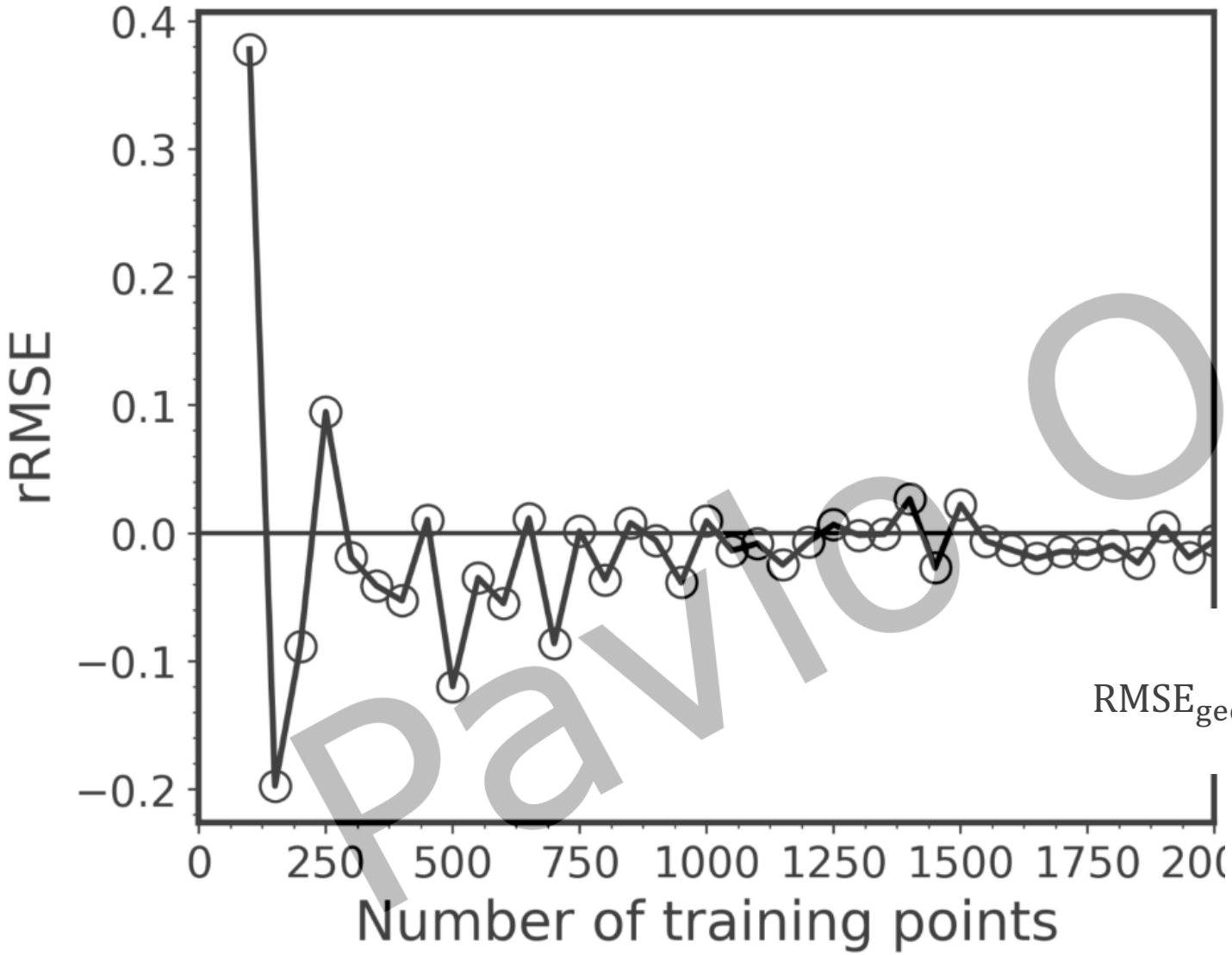
Train

The **KREG** model (**K**ernel ridge regression [KRR] with **RE** descriptor and the **G**aussian kernel function; RE descriptor stands for Internuclear distances **R**elative to **E**quilibrium) to complete all the ML tasks.

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P. O. Dral, A. Owens, S. Yurchenko, W. Thiel, *J. Chem. Phys.* **2017**, *146*, 244108

Automatic selection of training points



Optimal number of training points can be determined automatically using iterative procedure that stops after the relative change in ML validation errors drops below the threshold (typically rRMSE < 0.1).

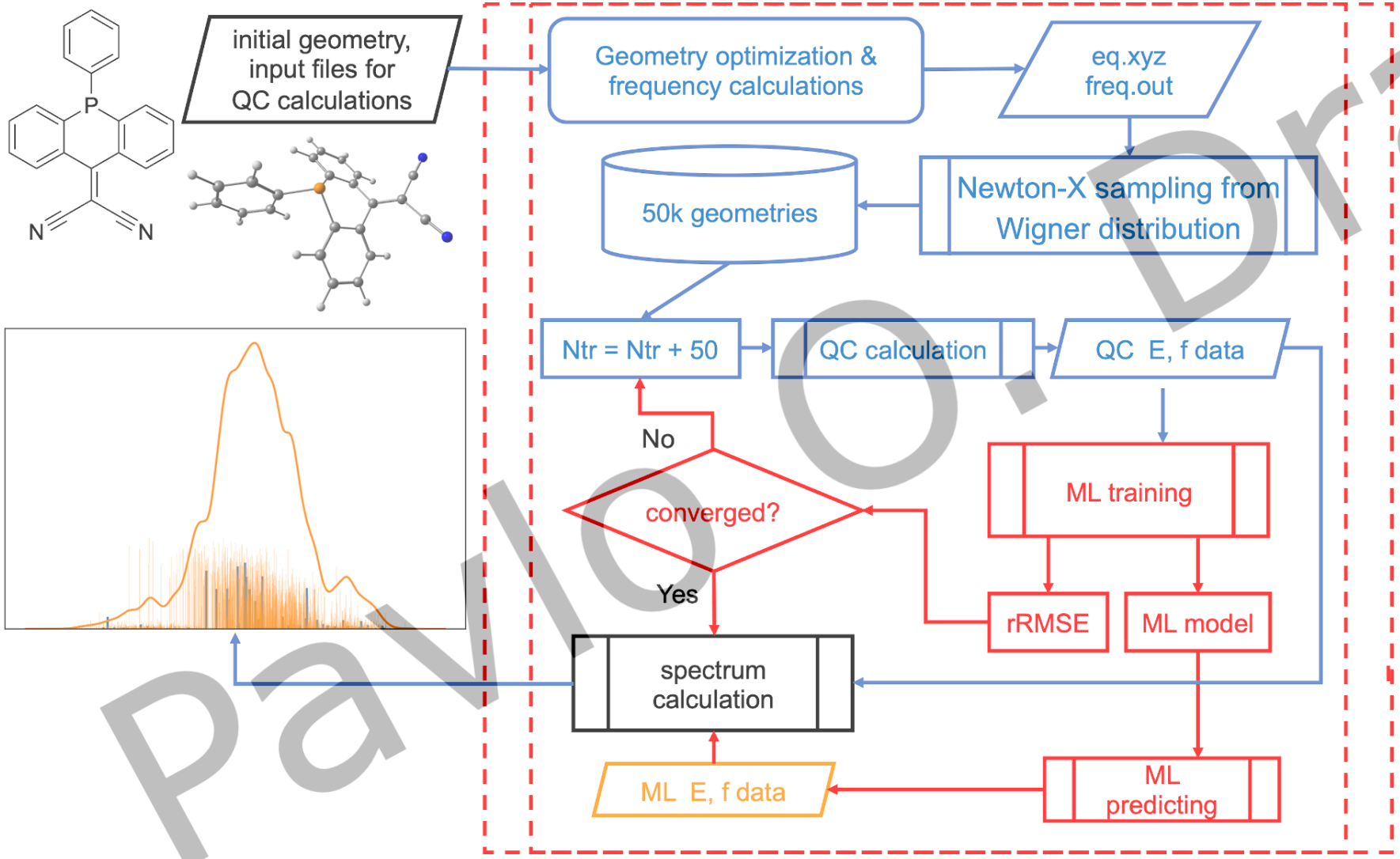
$$\mathrm{rRMSE} = \frac{RMSE_{geom}(N_{tr}) - RMSE_{geom}(N_{tr} - N_{step})}{RMSE_{geom}(N_{tr})}$$

$$\mathrm{RMSE}_{\mathrm{geom}}(N_{tr}) = \sqrt[2N_s]{\prod_{i=1}^{N_s} \mathrm{RMSE}_{\Delta E_{0n}}(N_{tr}) \cdot \mathrm{RMSE}_{f_{0n}}(N_{tr})}$$

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B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210

Machine learning single-photon absorption spectra





Mario Barbatti

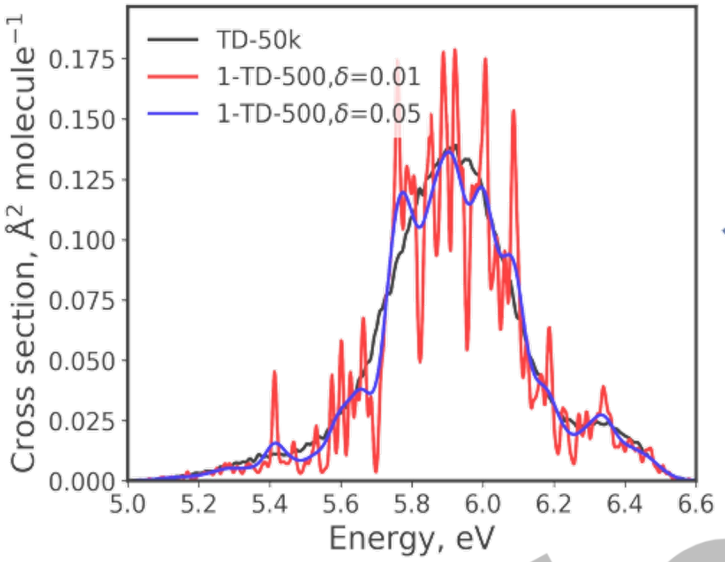


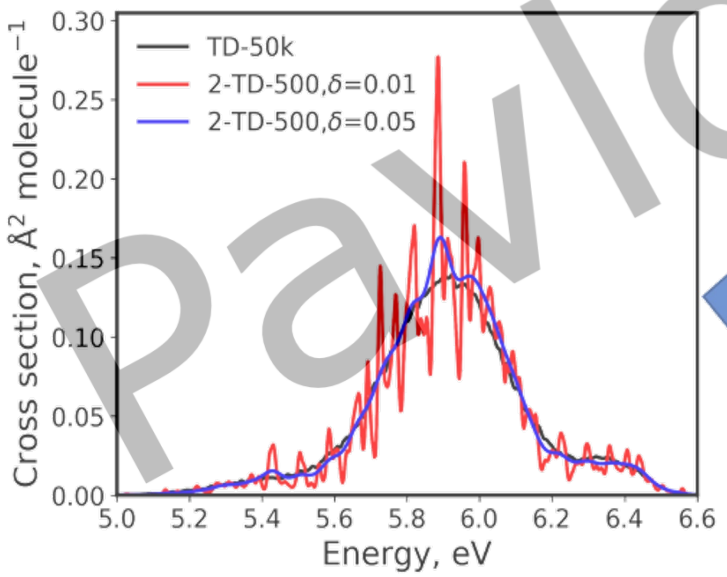
Bao-Xin Xue

Pavlo O. Dral

ML-NEA method: B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210
Implementation in MLatom: P. O. Dral, F. Ge, B.-X. Xue, Y.-F. Hou, M. Pinheiro Jr, J. Huang, M. Barbatti, *Top. Curr. Chem.*, ***2021***, *379*, 27

High-precision spectra of benzene

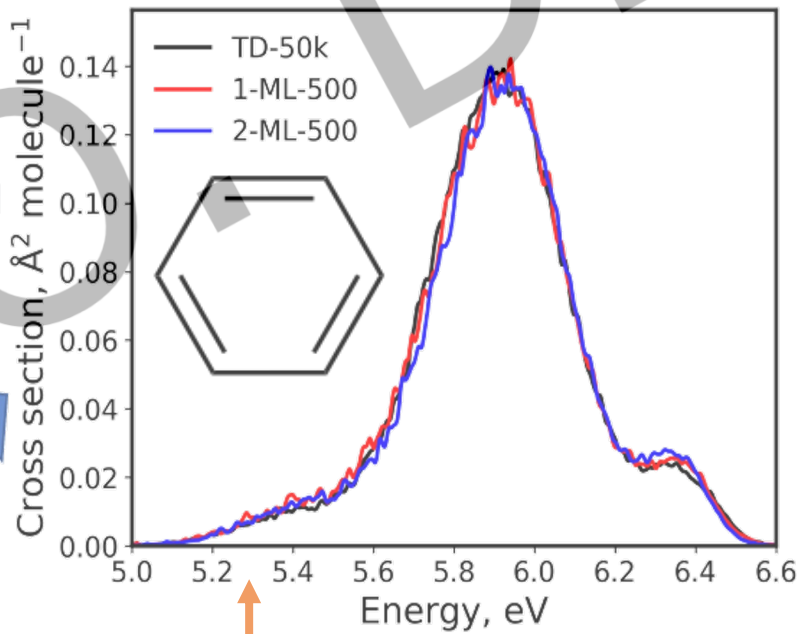




Two different mesium-sized ensembles of the same size (500 points) sampled from a Wigner distribution give very different spectra at QC level

train & predict

train & predict

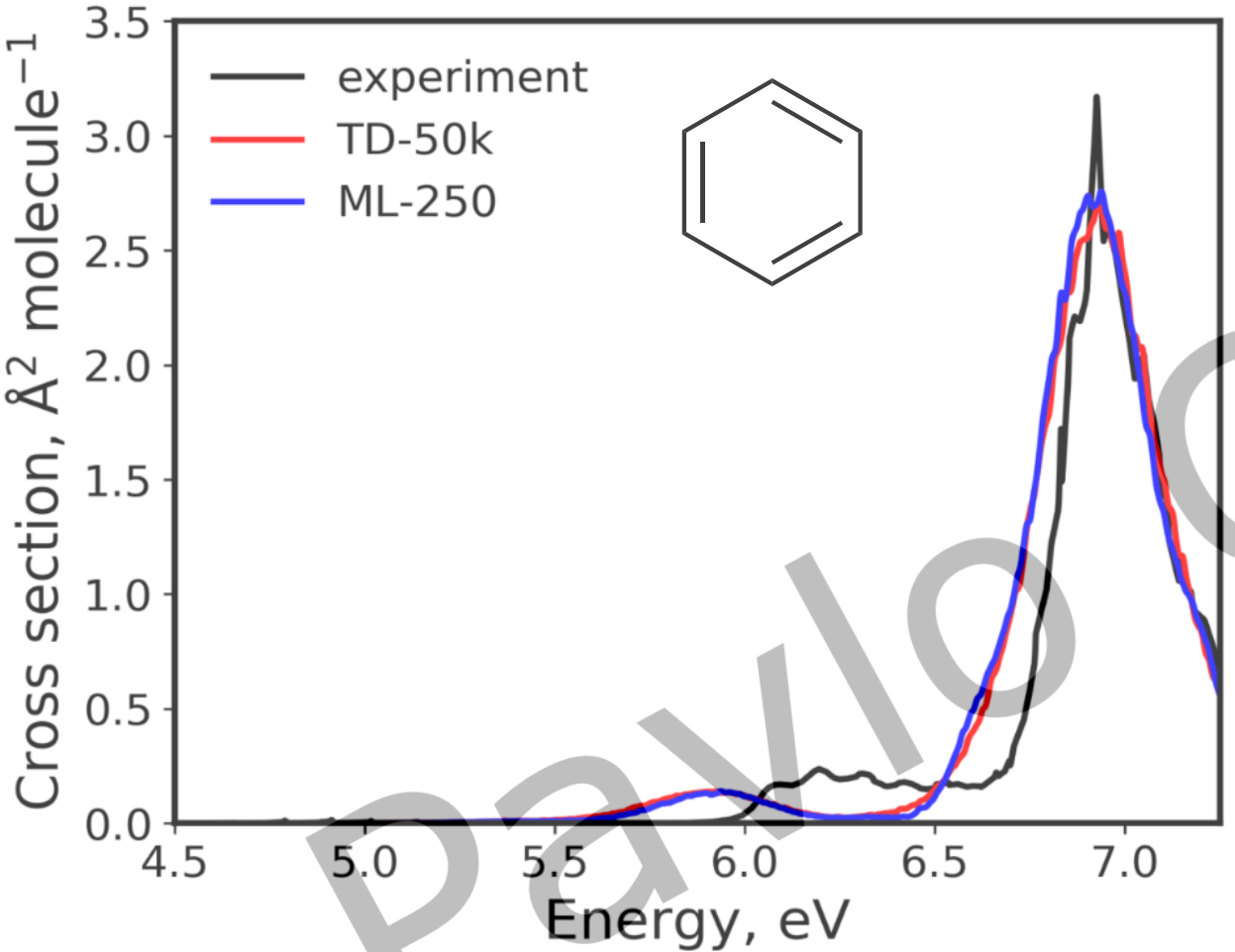


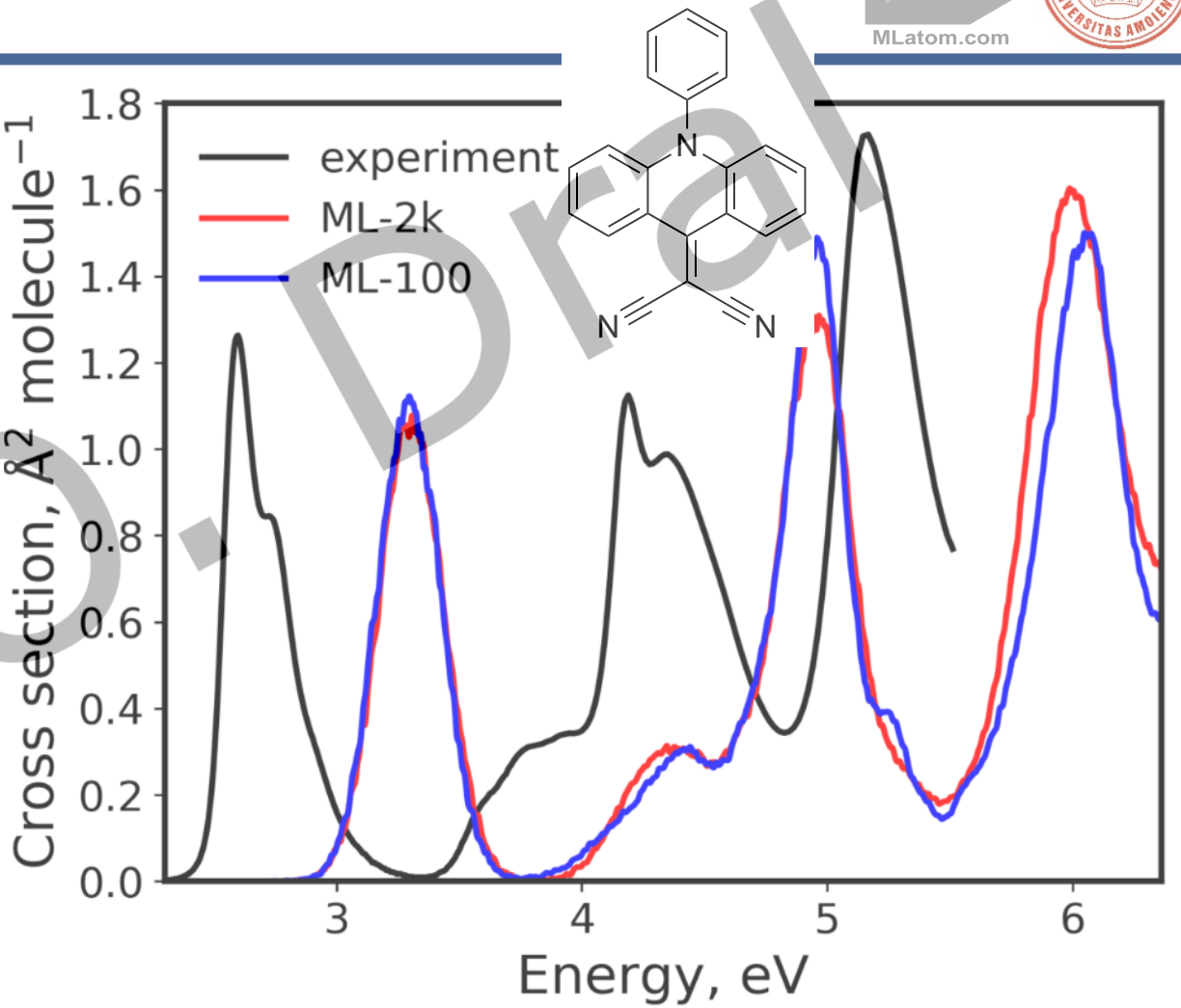
ML trained on both QC ensembles considerably improves precision of spectra that are very similar to each other

Pavlo O. Dral

B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210

The error of ML vs reference QM is smaller than the error of QM vs experiment

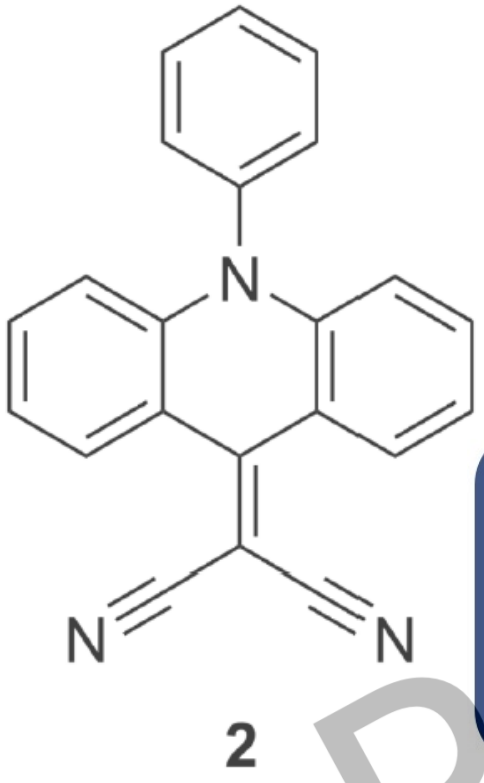




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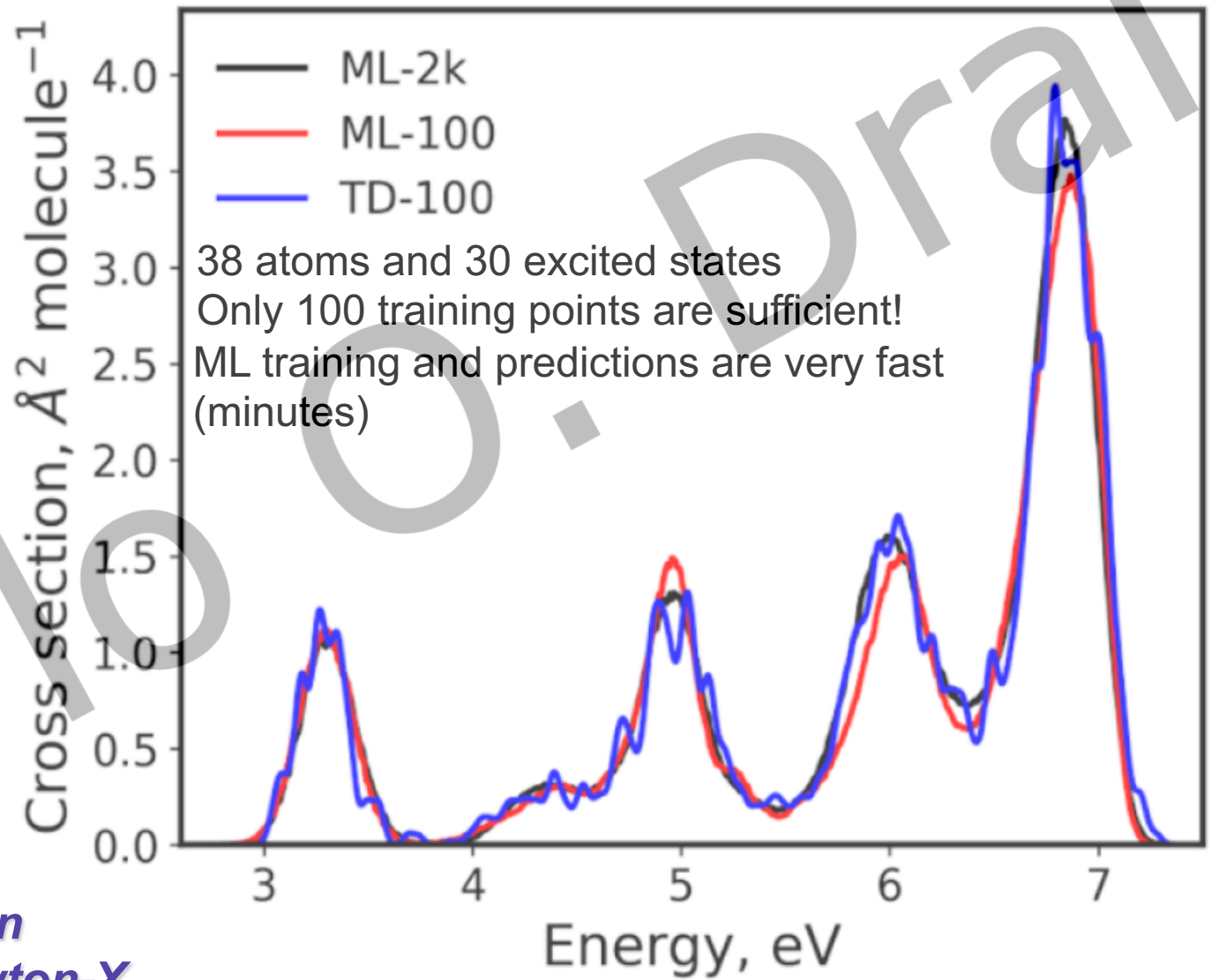
B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210

Black-box implementation into MLatom



MLatom.com

NX
newtonx.org

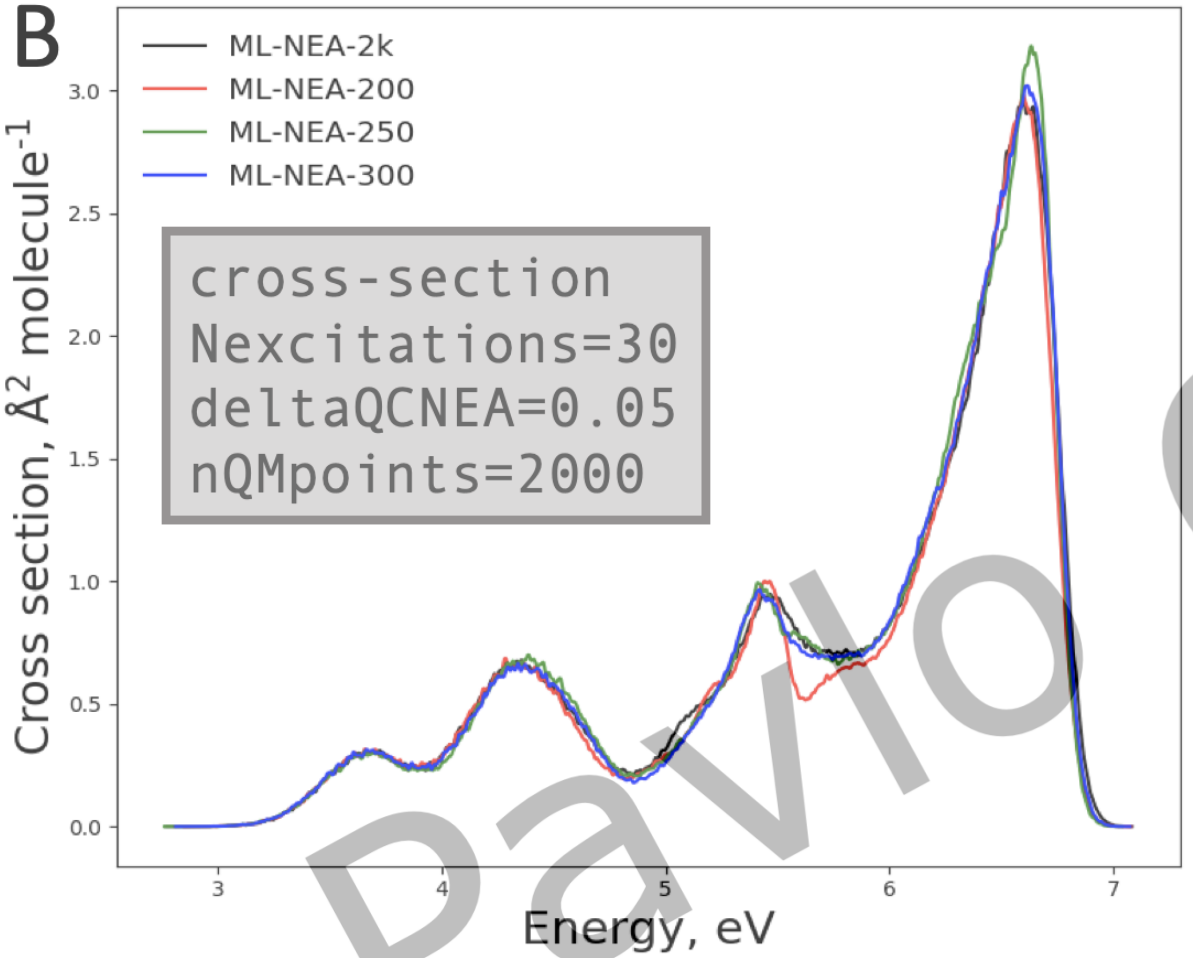


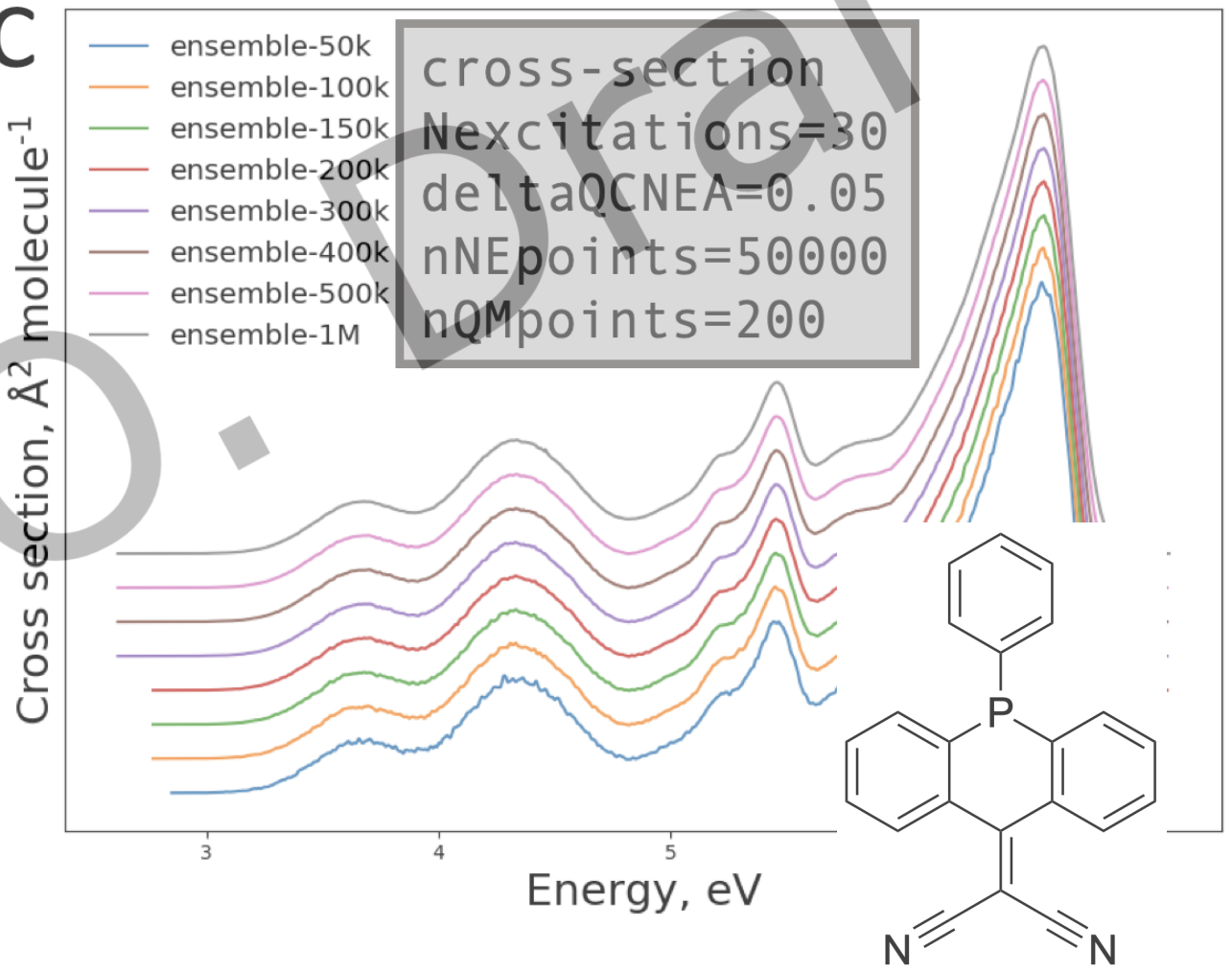
ML-NEA is available in MLatom interfaced to Newton-X

Pavlo O. Dral

B.-X. Xue, P. O. Dral, M. Barbatti, *J. Phys. Chem. A* **2020**, *124*, 7199–7210

Black-box implementation into MLatom





Pavlo O. Dral

Example ML-spectra-3.

The following task is from our book chapter.

MLatom input file:

```
cross-section
Nexcitations=30
plotQCNEA
plotQCSPC
deltaQCNEA=0.05
```

These calculations require many data files (reference excitation energies at TDDFT level). These data files are zipped. You should unzip them and upload all of them as auxiliary files to the cloud.

Create input file and submit the job in the File Manager.

Calculations can take more than 5 min. MLatom automatically determines the minimum required number of training points, in this case it needed 200 points for precise spectrum. In the output file you can find that it took 4 iterations to converge:

```
================================================================================
run ML-NEA iteratively for spectrum generation ( ML_train_iter ) started at Wed Dec  1 12:00:19 2021 CST
ML-NEA iteration 1: train_number = 50; RMSE_geom = 0.06717941145022376; rRMSE = 1.0

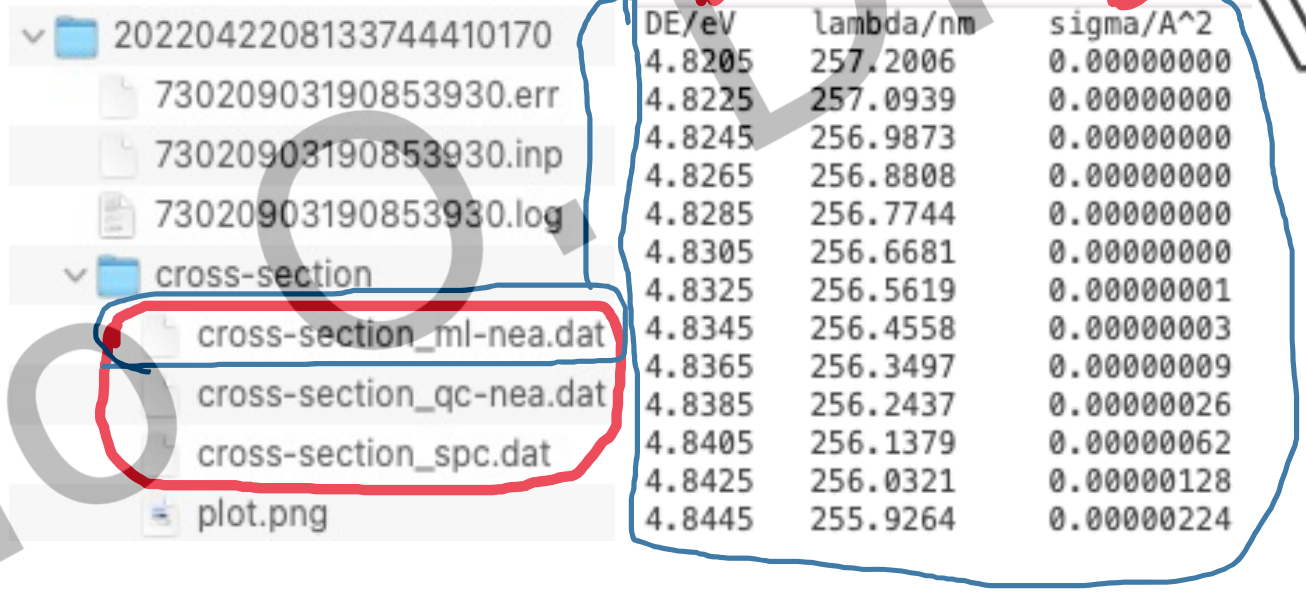
ML-NEA iteration 2: train_number = 100; RMSE_geom = 0.09043318436728051; rRMSE = 0.25713761026721255

ML-NEA iteration 3: train_number = 150; RMSE_geom = 0.06411060145373663; rRMSE = 0.410580813729204

ML-NEA iteration 4: train_number = 200; RMSE_geom = 0.0695737045717655; rRMSE = 0.07852252732055763

ML-NEA iteration ended after 4 iteration!
run ML-NEA iteratively for spectrum generation ( ML_train_iter ) finished at Wed Dec  1 12:08:01 2021 CST |||| total spent 462.02 sec
================================================================================
```

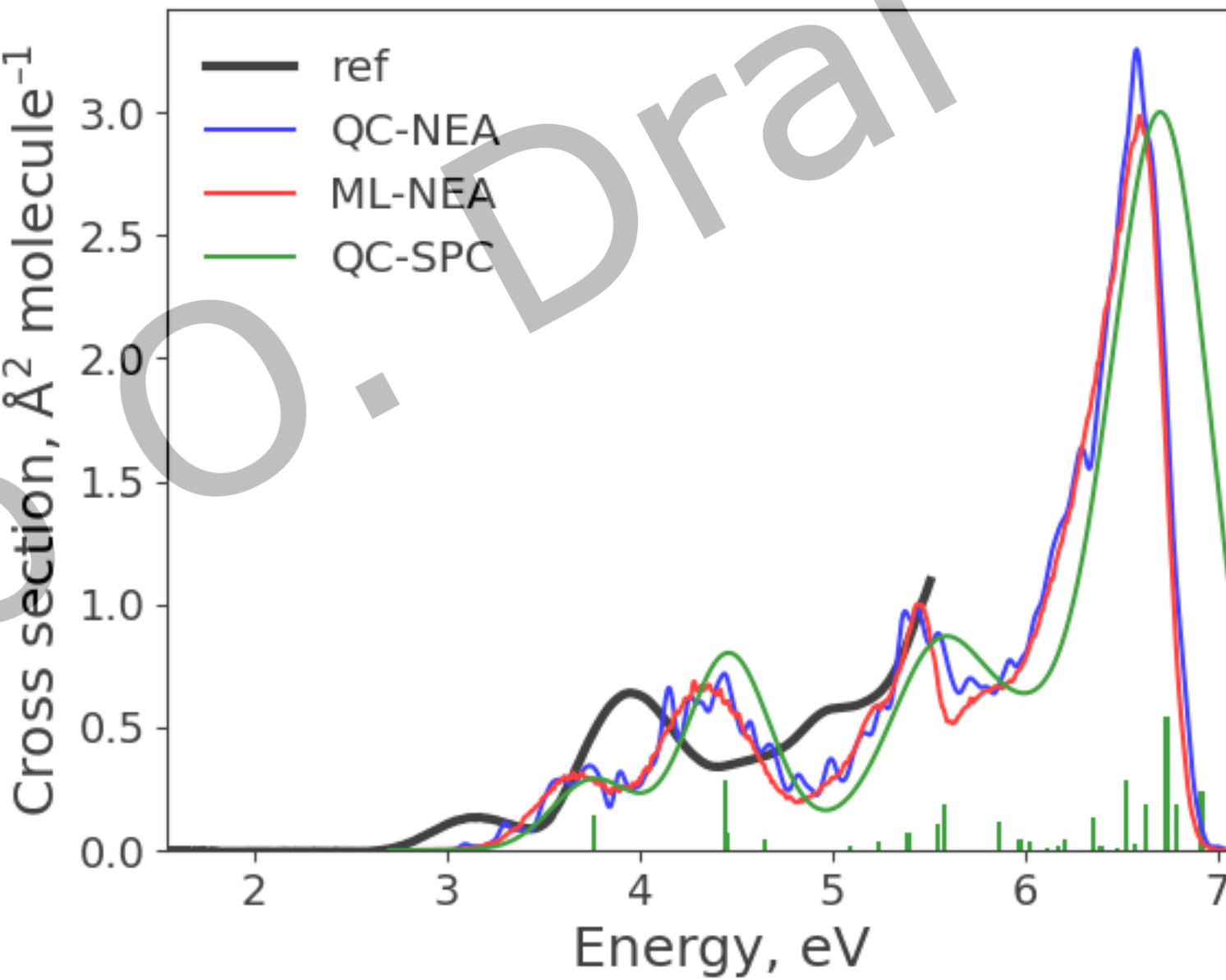
Pavlo O. Dral



Spectrum plotted for you in cross-section/plot.png

Note: You can also plot raw data from *.dat files with Origin or Excel

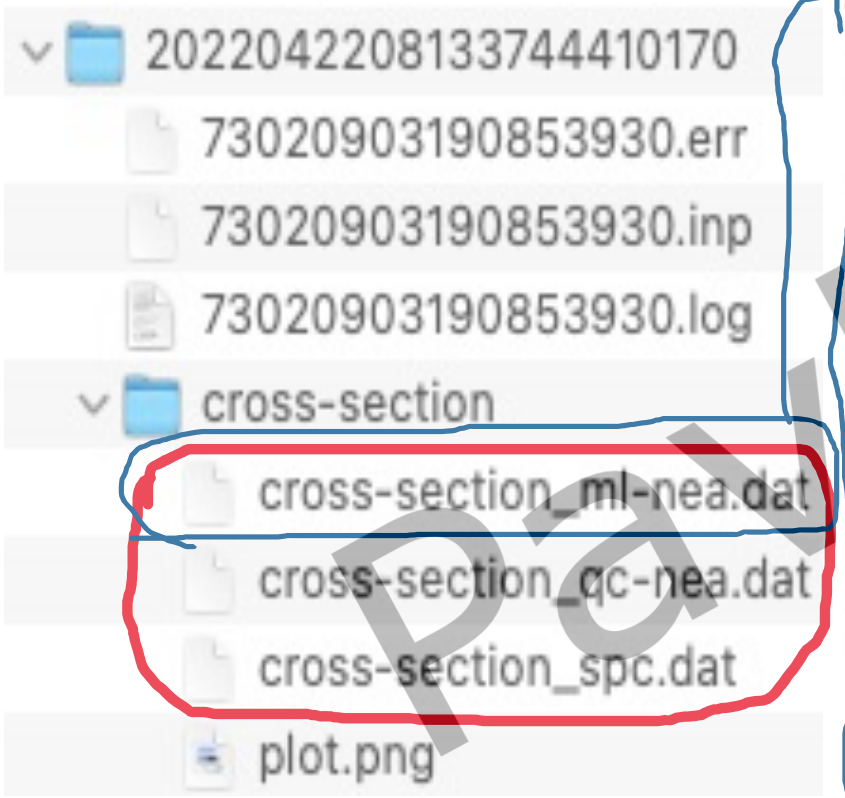
Pavlo O. Dral



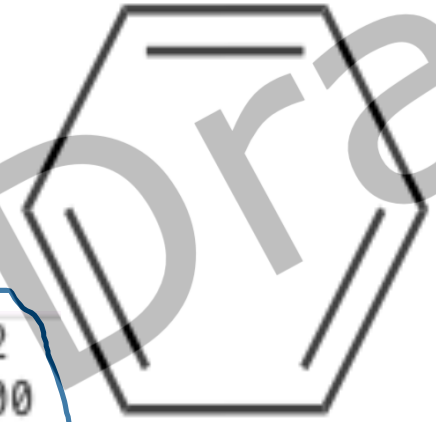
Spectrum plotted for you in cross-section/plot.png

Note: You can also plot raw data from *.dat files with Origin or Excel

Pavlo O. Dral



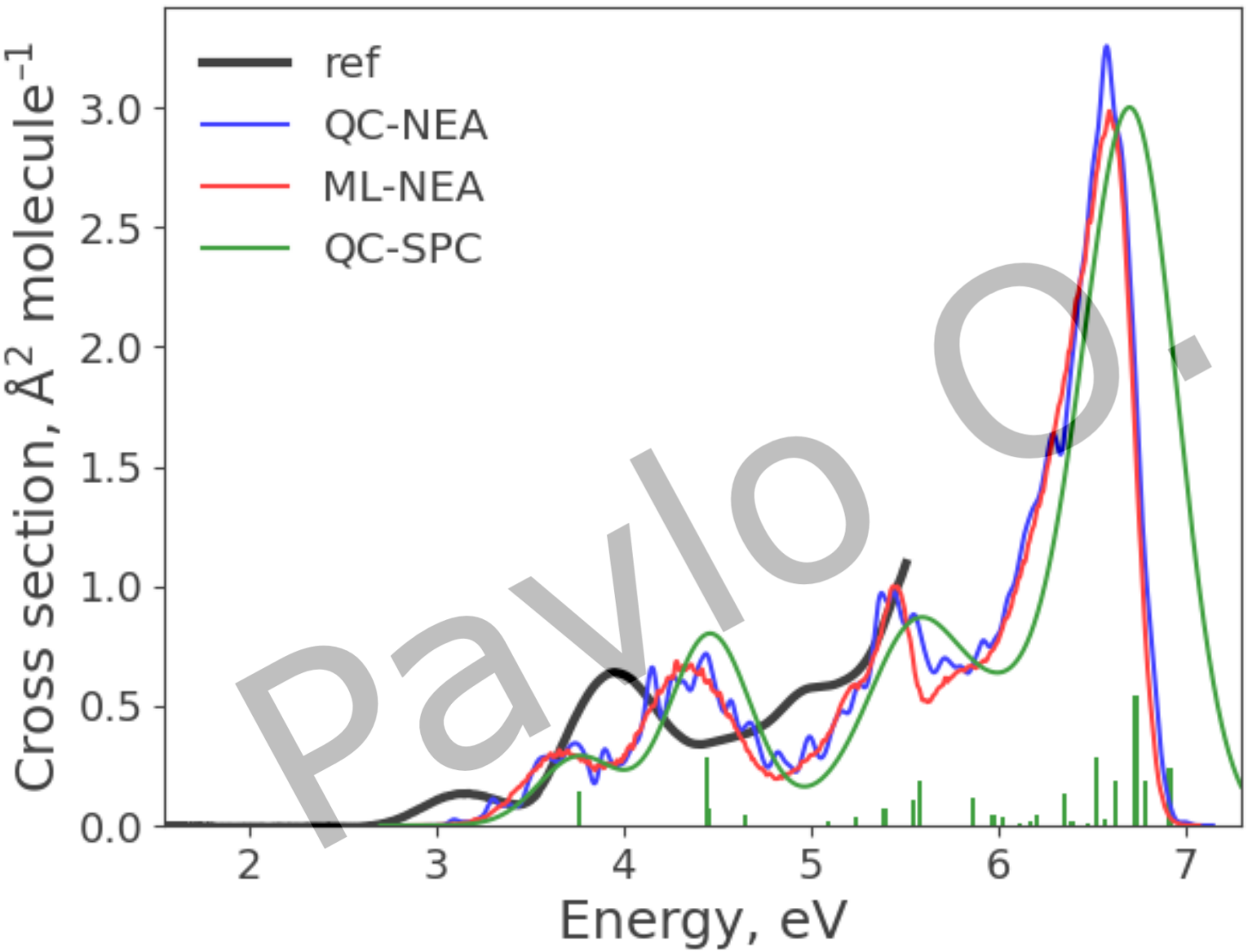
DE/eV	lambda/nm	sigma/A^2
4.8205	257.2006	0.00000000
4.8225	257.0939	0.00000000
4.8245	256.9873	0.00000000
4.8265	256.8808	0.00000000
4.8285	256.7744	0.00000000
4.8305	256.6681	0.00000000
4.8325	256.5619	0.00000001
4.8345	256.4558	0.00000003
4.8365	256.3497	0.00000009
4.8385	256.2437	0.00000026
4.8405	256.1379	0.00000062
4.8425	256.0321	0.00000128
4.8445	255.9264	0.00000224



Spectrum plotted for you in cross-section/plot.png

Note: You can also plot raw data from *.dat files with Origin or Excel

Payloo Dral



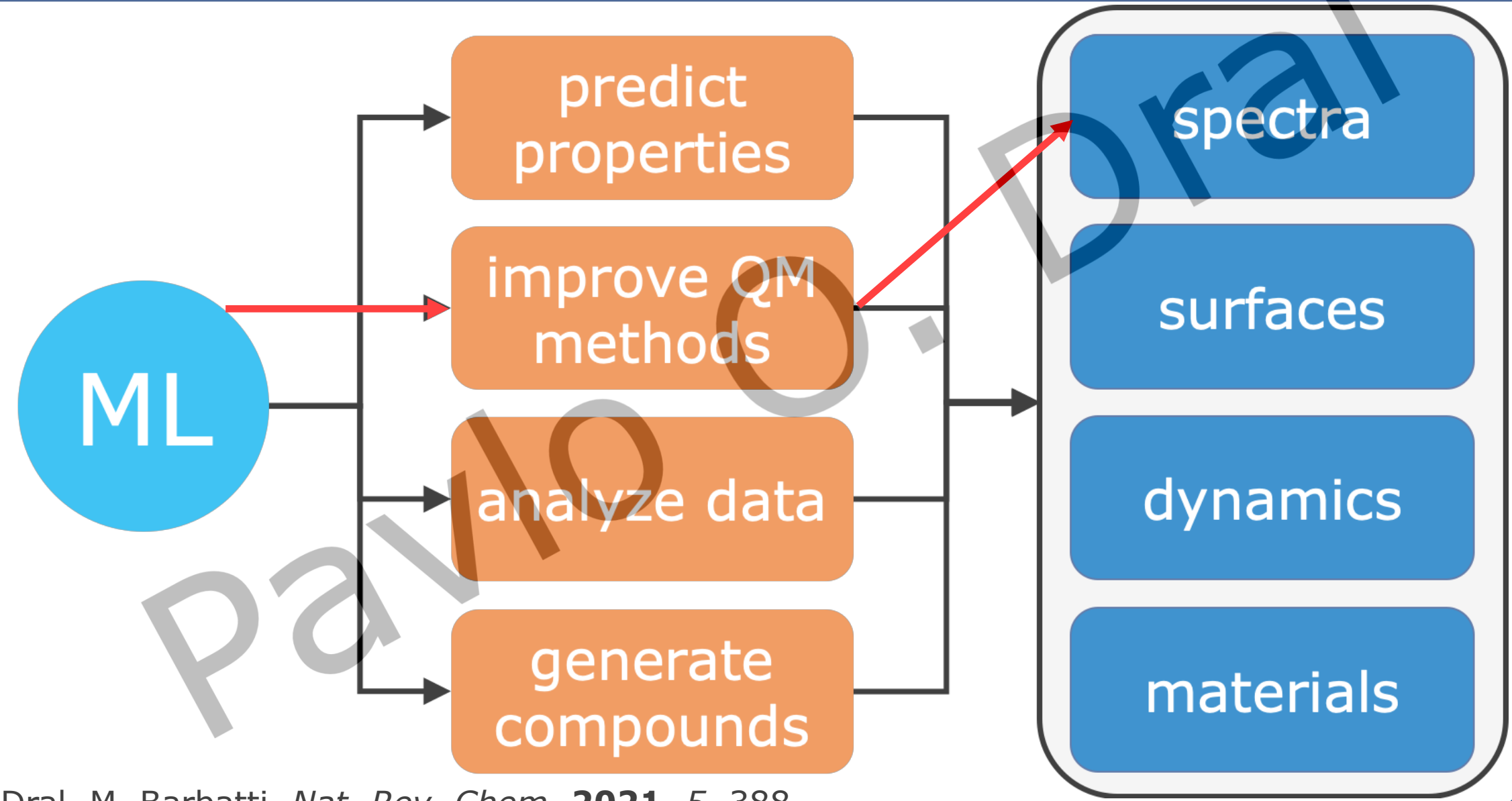


Spectrum plotted for you in cross-section/plot.png

Note: You can also plot raw data from *.dat files with Origin or Excel

Pavlo O. Dral

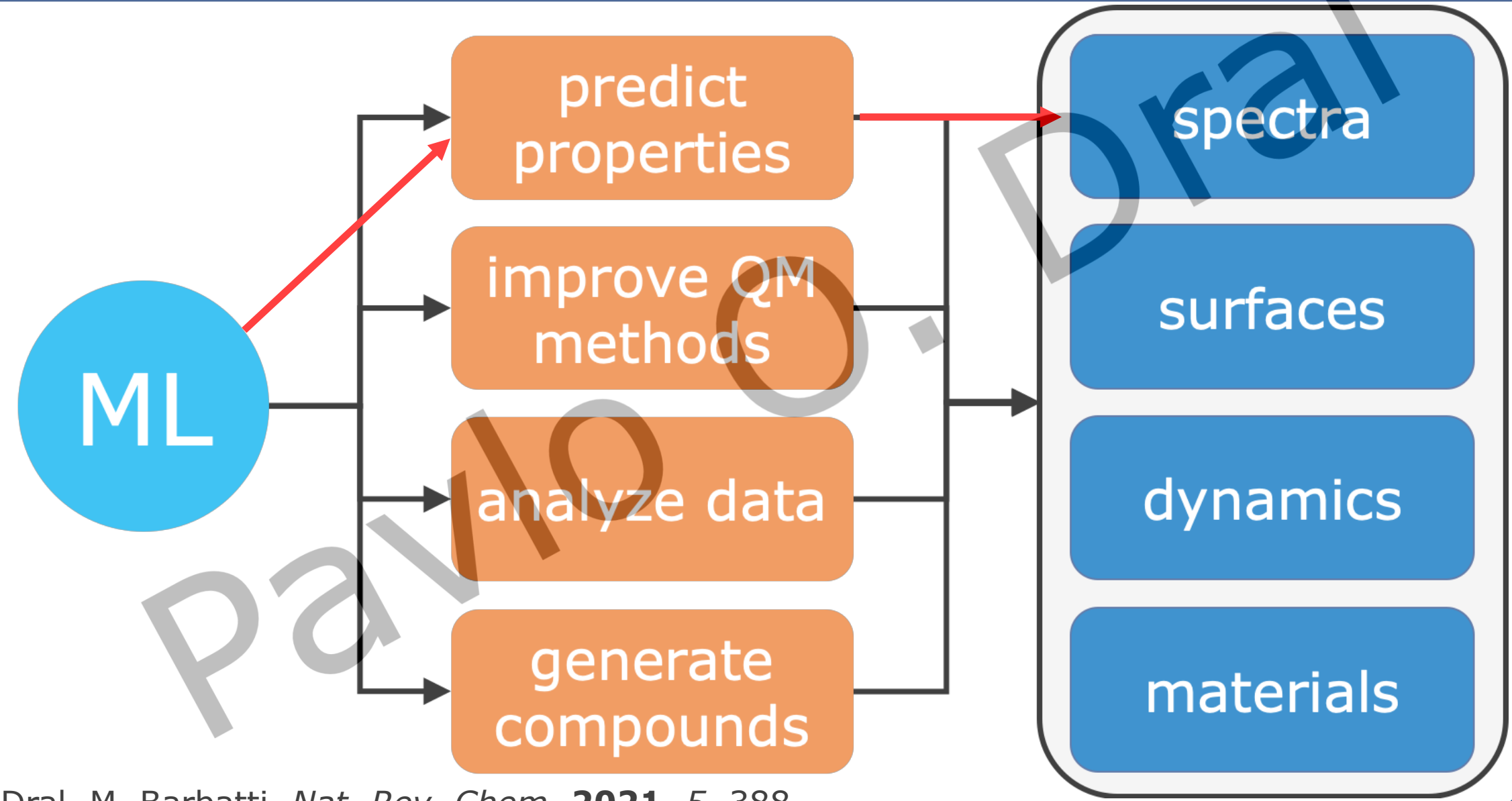
Machine learning in chemistry



Pavlo O. Dral

P. O. Dral, M. Barbatti, *Nat. Rev. Chem.* **2021,** *5*, 388

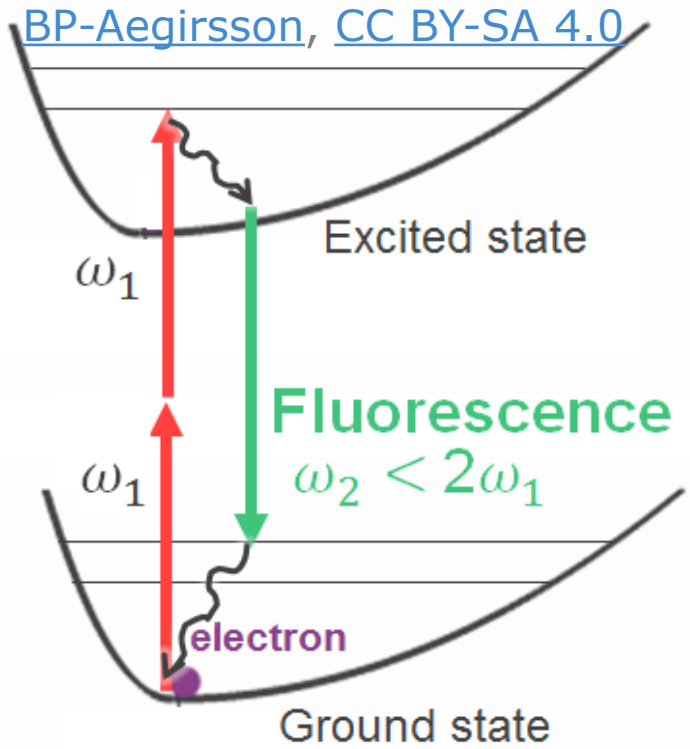
Machine learning in chemistry



Pavlo O. Dral

P. O. Dral, M. Barbatti, *Nat. Rev. Chem.* **2021,** *5*, 388

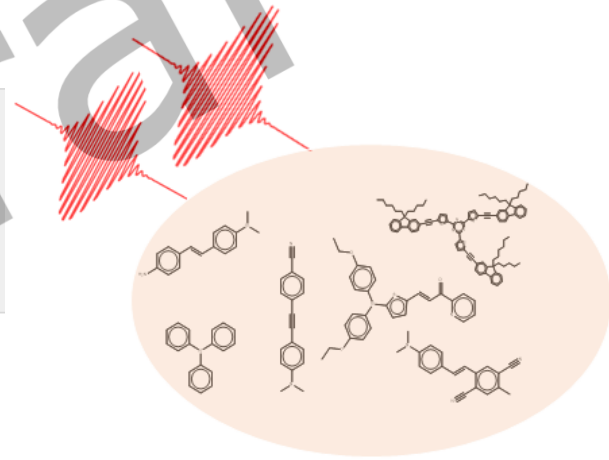
ML two-photon absorption



BP-Aegirsson, CC BY-SA 4.0

Here we show how to calculate TPA cross section for RHODAMINE 6G and RHODAMINE 123 molecules with MLatom input file `mltpa.inp`:

```
MLTPA
SMILESfile=Smiles.csv
auxfile=_aux.txt
```



This input requires `Smiles.csv` file with SMILES of molecules:

```
CCNC1=CC2=C(C=C1C)C(=C3C=C(C(=[NH+]CC)C=C302)C)C4=CC=CC=C4C(=O)OCC.[Cl-]
COC(=O)C1=CC=CC=C1C2=C3C=CC(=N)C=C30C4=C2C=CC(=C4)N.Cl
```

Two-photon absorption applications:

- two-photon lithography
- Photodynamic therapy
- Bioimaging
- 3D printing
- Upconverted laser


Yuming Su


Zhou Da


Cheng Wang

Pavlo O. Dral

Y. Su, Y. Dai, Y. Zeng, …, P. Zheng, D. Zhou, P. O. Dral, C. Wang. *Adv. Sci.* **2023**, 2204902

Two-photon absorption

Example ML-spectra-4.

Taking an example from MLatom's documentation, the input file to calculate the TPA of RHODAMINE 6G and RHODAMINE 123 molecules:

```
MLTPA
SMILESfile='
CCNC1=CC2=C(C=C1C)C(=C3C=C(C(=[NH+]CC)C=C3O2)C)C4=CC=CC=C4C(=O)OCC.[Cl-]
COC(=O)C1=CC=CC=C1C2=C3C=CC(=N)C=C3OC4=C2C=CC(=C4)N.Cl
'
auxfile='
600,850,55.4
600,600,33.9
'
```

Wavelength_lower_bound, wavelength_upper_bound, Et30 (defines solvent)

After the calculations finish, the predicted TPA cross section values are saved in a folder named `tpa[absolute time]`. In the folder, there are two files for two molecules: `tpa1.txt` and `tpa2.txt`.

Analyze and plot the spectrum in tpa1.txt.

How long time did you take you to peform calculations?

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Y. Su, Y. Dai, Y. Zeng, …, P. Zheng, D. Zhou, P. O. Dral, C. Wang. *Adv. Sci.* **2023**, 2204902

Two-photon absorption

After the calculations finish, the predicted TPA cross section values are saved in two files for two molecules: `tpa1.txt` and `tpa2.txt`. For our examples, they look like:

tpa1.txt

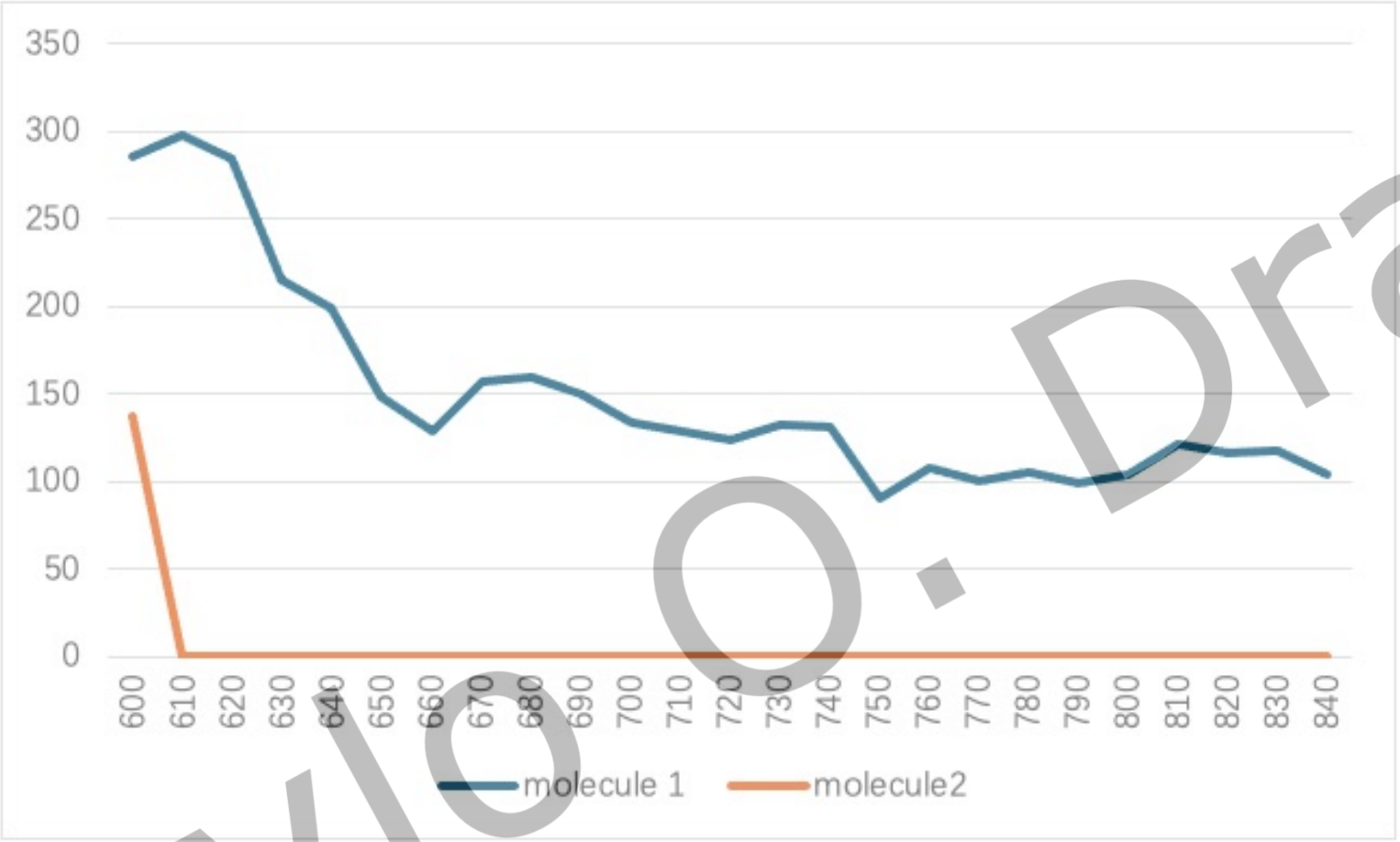
```
wavelength,predicted_sigma (GM)
600.0,285.19455
610.0,297.71707
620.0,284.11694
......
810.0,121.51988
820.0,116.537994
830.0,118.04909
840.0,103.65925
850.0,113.72374
```

tpa2.txt

```
wavelength,predicted_sigma (GM)
600.0,138.2346
```

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Y. Su, Y. Dai, Y. Zeng, …, P. Zheng, D. Zhou, P. O. Dral, C. Wang. *Adv. Sci.* **2023**, 2204902



Time used :Wall-clock time: 2.41 s

Molecule 1 has much stronger TPA than molecule 2, and TPA weakens As the wavelength increases.

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