[1] Long timescale surface hopping dynamics with deep NNs

**SHARC with multi-layer feed-forward NNs**
- Use NNs instead of QM calculations to predict energies, gradients, couplings and dipole moments
- Relationships between the nuclear coordinates and the corresponding electronic properties are learned from a training set

**Training set generation and adaptive sampling for excited states**
- Each data point is one set of nuclear coordinates and its associated set of quantities computed with a reference method.
- Initial training set based on normal mode scans and then switch to an adaptive sampling scheme that automatically identifies untrustworthy regions not covered by the initial training set.
- Whenever the different NNs make different predictions, the corresponding geometry is assumed to lie in a conformational region with too few training points → expand the training set by computing the quantum chemistry data for this geometry.
- During the run, threshold for the error between the NNs is adapted (x0.95) until the conformational space is sampled to make accurate predictions without any reference calculation.

**Results for CH$_3$NH$_2$**
- MAE of 0.057 and 0.099 between NNs and two reference methods (MR-CISD with different b.s.).
- Very low comp. cost, so a much larger number of trajectories (3,846) was simulated than with standard QM methods (90).
- Long time scale excited-state dynamics (up to 1 ns, 10$^4$ times longer than QM).
- 10 ps trajectory in less than 6 hours on 1 core (300x faster than ref. QM).
- Correct identification of the Cis.

[2] Pure deep learning vs. ab initio non-adiabatic excited state dynamics

**Feed-forward deep NNs with Zhu-Nakamura TSH method for pure ML non-adiabatic dynamics**
- Aim: explore topology of S$_0$ and S$_1$ PESs and the S$_0$/S$_1$ conical intersection of CH$_3$NH.
- DNNs model where PES of the molecule is expressed as sum of energies of constituent atoms.

\[ E = \sum_i E_i \quad \text{with} \quad E_i = F_i^{\text{out}} \left( F_i^{\text{out}} \left( \sum_j F_j^{\text{in}} (\{d_j^i\}) \right) \right) \]

Two independent DNN models trained independently for both S$_0$ and S$_0$ PESs of CH$_3$NH on CASSCF data calculated for the same set of molecular coordinates.

Final DNN models are trained on 90,000 ab initio data points prepared using molecular dynamics simulations.

S$_0$ and S$_1$ energy profiles calculated from the trained DNN models fully overlap with CASSCF. S$_1$/S$_0$ CI topology well reproduced (see 2D and 3D PES ⇨)

**Results for DNNs**
- S$_1$-S$_0$ gap same well reproduced at all hopping and Franck-Condon regions
- TD S$_1$ and S$_0$ populations match ab-initio ones
- S$_1$ to S$_0$ hopping starts at same time (55 fs)
- Similar distribution of hopping points