

# **Supplementary Materials: Nonadiabatic Derivative Couplings Calculated Using Information of Potential Energy Surfaces without Wavefunctions: Ab Initio and Machine Learning Implementations**

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## 1. Additional Data

**Table S1.** Number of structures for  $\Delta E_{S_0S_1}$  in different groups.

Energy Gap (kcal/mol)	Number of Data
0.17 (MECI)	1
0.17-1	5
1-3	42
3-5	85
5-10	379
10-15	319
> 15	130

**Table S2.** Root mean square deviation (RMSD) values (in Bohr<sup>-1</sup>) of the PES-based NACMEs with respect to the wavefunction-based NACMEs.

Energy Gap (kcal/mol)	RMSD (Bohr <sup>-1</sup> )
0.17 (MECI)	86.09
0.17-1	36.71
1-3	2.24
3-5	1.15
5-10	0.66
10-15	0.41
> 15	0.27

**Table S3.** Root mean square deviation (RMSD) values (in Bohr<sup>-1</sup>) of the SA-CASSCF- and ML-calculated NACMEs (PES-based algorithm).

Energy Gap (kcal/mol)	RMSD (Bohr <sup>-1</sup> )
0.17 (MECI)	46.79
0.17-1	33.71
1-3	6.09
3-5	1.46
5-10	0.40
10-15	0.19
> 15	0.17

## 2. Cartesian Coordinates of the optimized CI structure (In Å)

C	0.6469	-0.0000	0.0502
H	1.1917	0.9240	-0.0041
H	1.1888	-0.9254	-0.0045
N	-0.7133	0.0002	-0.1428
H	-1.2689	-0.0002	0.7074

## 3. Branching space vectors at the optimized CI structure

**g**

-0.0547998078	-0.0289750951	-0.0267761876
0.0005912536	-0.0007005861	-0.0326824556
0.0037434127	-0.0011734331	0.0284251728
0.0631510096	0.0975623138	0.0485796303
-0.0126858681	-0.0667131995	-0.0175461599

**h**

-0.0619946799	0.0134129868	-0.0074055181
0.0073314251	-0.0005031829	0.0067776450
0.0029965689	-0.0022956584	-0.0123248759
0.0632778457	-0.0270384598	0.0279724305
-0.0116111599	0.0164243143	-0.0150196815