



UNIVERSITY OF LEEDS

This is a repository copy of *Nonadiabatic Excited-State Molecular Dynamics Methodologies: Comparison and Convergence*.

White Rose Research Online URL for this paper:
<https://eprints.whiterose.ac.uk/172025/>

Version: Supplemental Material

Article:

Freixas, VM, White, AJ, Nelson, T et al. (5 more authors) (2021) Nonadiabatic Excited-State Molecular Dynamics Methodologies: Comparison and Convergence. *Journal of Physical Chemistry Letters*, 12 (11). pp. 2970-2982. ISSN 1948-7185

<https://doi.org/10.1021/acs.jpcclett.1c00266>

Reuse

Items deposited in White Rose Research Online are protected by copyright, with all rights reserved unless indicated otherwise. They may be downloaded and/or printed for private study, or other acts as permitted by national copyright laws. The publisher or other rights holders may allow further reproduction and re-use of the full text version. This is indicated by the licence information on the White Rose Research Online record for the item.

Takedown

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing eprints@whiterose.ac.uk including the URL of the record and the reason for the withdrawal request.



eprints@whiterose.ac.uk
<https://eprints.whiterose.ac.uk/>

Supporting Information for

Non-Adiabatic Excited State Molecular Dynamics Methodologies:

Comparison and Convergence

*Victor M. Freixas,¹ Alexander J. White,² Tammie Nelson,² Huajing Song,² Dmitry V. Makhov,^{3,4}
Dmitrii Shalashilin,³ Sebastian Fernandez-Alberti,^{*1} Sergei Tretiak^{*2}*

- 1- Universidad Nacional de Quilmes, Roque Saénz Peña 352, B1876BXD Bernal, Argentina
- 2- Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
- 3- School of Chemistry, University of Leeds, Leeds, LS2 9JT, UK
- 4- School of Mathematics, University of Bristol, Bristol BS8 1TW, UK

AUTHOR INFORMATION

Corresponding Author

* E-mail: sfalberti@gmail.com, serg@lanl.gov

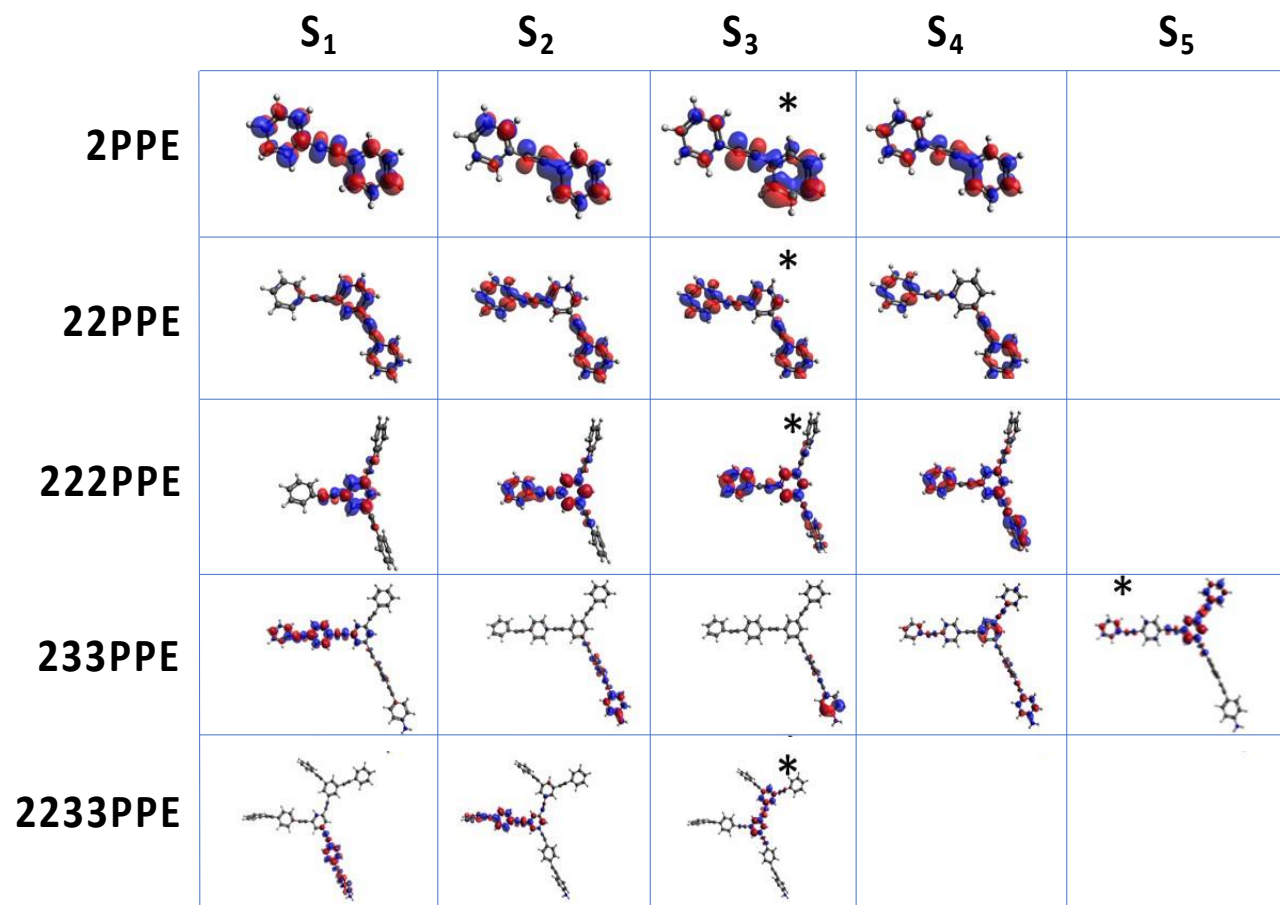


Figure S1. Localization of electronic transition densities for the excited states considered in our simulations for a random initial configuration from the 300 K conformational sampling. The asterisk marks the initial state for excited state dynamics.

Table S1. AIMC parameters used to generate data presented in **Figure 5**.

Molecule	panel	Number of initial conditions	δ_2 (cloning criteria)	Maximum number of cloning events allowed	Gaussian widths	Total number of cloning events
2PPE	a)	100	5°	16	α	980
2PPE	a)	200	5°	16	α	2550
2PPE	a)	300	5°	16	α	4110
2PPE	a)	400	5°	16	α	5676
2PPE	a)	500	5°	16	α	7269
2PPE	a)	600	5°	16	α	8822

222PPE	b)	100	5°	16	α	1072
222PPE	b)	200	5°	16	α	2750
222PPE	b)	300	5°	16	α	4312
222PPE	b)	400	5°	16	α	6065
222PPE	b)	500	5°	16	α	7728
222PPE	b)	600	5°	16	α	9376
2PPE	c)	300	15°	16	α	2942
2PPE	c)	300	10°	16	α	3674
2PPE	c)	300	5°	16	α	4110
2PPE	c)	300	1°	16	α	4162
2PPE	c)	300	5°	32	α	7936
2PPE	c)	300	1°	32	α	8070
222PPE	d)	300	15°	16	α	3077
222PPE	d)	300	10°	16	α	4098
222PPE	d)	300	5°	16	α	4312
222PPE	d)	300	1°	16	α	4346
222PPE	d)	300	5°	32	α	8954
222PPE	d)	300	1°	32	α	9087
2PPE	e)	300	5°	16	0 α	4110
2PPE	e)	300	5°	16	0.5α	4110
2PPE	e)	300	5°	16	α	4110
2PPE	e)	300	5°	16	2α	4312
222PPE	f)	300	5°	16	0 α	4312
222PPE	f)	300	5°	16	0.5α	4312
222PPE	f)	300	5°	16	α	4312
222PPE	f)	300	5°	16	2α	4312

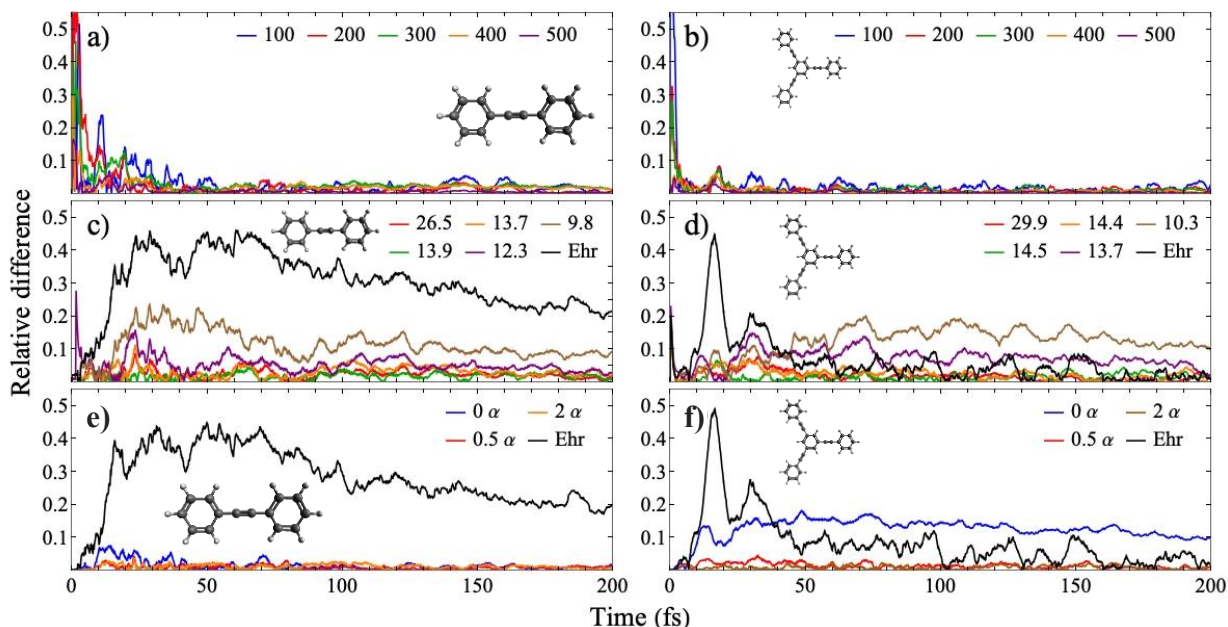


Figure S2. The relative differences between the S_1 adiabatic state populations obtained from EHR and AIMC simulations using different parameters. Relative differences between populations are calculated with respect to our most accurate result as $abs(a-b)/mean(a,b)$ where a is our tested parameter and b is our most accurate result. (a) and (b) Convergence of AIMC results with respect to the number of trajectories for 2PPE and 222PPE, respectively. Relative difference with respect to a 600 trajectory ensemble. (c) and (d) Convergence of AIMC results with respect to the number of cloning events per initial condition for 2PPE and 222PPE, respectively. Relative difference with respect to largest number of cloning events (26.9 for 2PPE and 30.3 for 222PPE). (e) and (f) Analysis of robustness of AIMC results with respect to variations in the α Gaussian width for 2PPE and 222PPE, respectively. Relative error with respect to 1α .