Supporting Information for: Probing Dynamics in Higher-Lying Electronic States with Resonance-Enhanced Femtosecond Stimulated Raman Spectroscopy

Timothy J. Quincy, Matthew S. Barclay, Marco Caricato,* and Christopher G. Elles*

Department of Chemistry, University of Kansas, Lawrence, Kansas 66045

E-mail: mcaricato@ku.edu; elles@ku.edu

This document includes tables of all optimized geometries and calculated vibrational frequencies for S_0 and S_1, as well as the calculated off-resonance Raman intensities, transition strengths for $S_n \leftarrow S_1$ excitations, and calculated S_n gradients. Figures also show the displacement vectors for several key S_1 vibrations; compare experimental ground- and excited-state absorption spectra with the calculated spectra for S_0 and S_1, respectively; and show the geometry and basis set dependence for simulated excited-state resonance Raman spectra using EOM-CCSD.

Table S1: Ground-state (S_0) geometries for the optimized conformers of DPT belonging to C_2, C_S, and C_{2v} point groups (B3LYP/aug-cc-pVDZ). The C_S conformation is the global minimum, with relative energies of 0.0038 eV for C_2 and 0.0072 eV for C_{2v}.

Atom	C_2			C_S			C_{2v}		
	Cartesian Coordinates (Å)			Cartesian Coordinates (Å)			Cartesian Coordinates (Å)		
	X	Y	Z	X	Y	Z	X	Y	Z
S	0.000000	0.000000	0.720985	-0.700102	-0.186412	0.000000	0.000000	0.000000	0.717566
C	0.000276	1.270819	-0.492294	0.475081	0.116620	1.270166	0.000000	1.272867	-0.494373
C	0.001171	0.710319	-1.753734	1.699982	0.419222	0.710466	0.000000	0.709607	-1.755350
C	-0.001171	-0.710319	-1.753734	1.699982	0.419222	-0.710466	0.000000	-0.709607	-1.755350
C	-0.000276	-1.270819	-0.492294	0.475081	0.116620	-1.270166	0.000000	-1.272867	-0.494373
C	0.000000	2.690128	-0.113915	0.107979	0.028389	2.689396	0.000000	2.692102	-0.115133
C	0.000000	-2.690128	-0.113915	0.107979	0.028389	-2.689396	0.000000	-2.692102	-0.115133
C	0.418261	3.115079	1.162660	-1.224263	0.185405	3.120129	0.000000	3.097582	1.234502
C	0.412532	4.467219	1.507934	-1.555939	0.102502	4.473244	0.000000	4.448881	1.582301
C	-0.011167	5.430367	0.587044	-0.564856	-0.133663	5.431039	0.000000	5.434266	0.590542
C	-0.429479	5.023603	-0.684815	0.762775	-0.291860	5.018155	0.000000	5.048655	-0.754743
C	-0.424566	3.672747	-1.032016	1.095393	-0.217912	3.665580	0.000000	3.698871	-1.103733
C	0.424566	-3.672747	-1.032016	1.095393	-0.217912	-3.665580	0.000000	-3.698871	-1.103733
C	0.429479	-5.023603	-0.684815	0.762775	-0.291860	-5.018155	0.000000	-5.048655	-0.754743
C	0.011167	-5.430367	0.587044	-0.564856	-0.133663	-5.431039	0.000000	-5.434266	0.590542
C	-0.412532	-4.467219	1.507934	-1.555939	0.102502	-4.473244	0.000000	-4.448881	1.582301
C	-0.418261	-3.115079	1.162660	-1.224263	0.185405	-3.120129	0.000000	-3.097582	1.234502
H	0.014870	1.303224	-2.665862	2.575798	0.671049	1.305042	0.000000	1.296859	-2.670803
H	-0.014870	-1.303224	-2.665862	2.575798	0.671049	-1.305042	0.000000	-1.296859	-2.670803
H	0.771258	2.381914	1.888163	-2.007153	0.392371	2.390259	0.000000	2.348772	2.026701
H	0.741310	4.769259	2.502855	-2.594219	0.230306	4.781257	0.000000	4.732417	2.635265
H	-0.015635	6.486620	0.856782	-0.824050	-0.196592	6.488091	0.000000	6.490137	0.861606
H	-0.762397	5.764440	-1.412660	1.543789	-0.486091	5.754165	0.000000	5.805953	-1.539402
H	-0.767624	3.375669	-2.022339	2.129742	-0.371963	3.360167	0.000000	3.426396	-2.157961
H	0.767624	-3.375669	-2.022339	2.129742	-0.371963	-3.360167	0.000000	-3.426396	-2.157961
H	0.762397	-5.764440	-1.412660	1.543789	-0.486091	-5.754165	0.000000	-5.805953	-1.539402
H	0.015635	-6.486620	0.856782	-0.824050	-0.196592	-6.488091	0.000000	-6.490137	0.861606
H	-0.741310	-4.769259	2.502855	-2.594219	0.230306	-4.781257	0.000000	-4.732417	2.635265
H	-0.771258	-2.381914	1.888163	-2.007153	0.392371	-2.390259	0.000000	-2.348772	2.026701

Table S2: Calculated ground-state (S_0) vibrational frequencies and Raman activities for the three conformers of DPT (B3LYP/aug-cc-pVDZ).

C_2				C_S				C_{2v}			
Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry
ν_1	20	13	a	ν_1	27	0	a"	ν_1	12	0	b_1
ν_2	27	0	b	ν_2	34	13	a'	ν_2	14	0	a_2
ν_3	51	1	b	ν_3	53	7	a'	ν_3	52	0	b_1
ν_4	69	4	a	ν_4	70	6	a'	ν_4	77	3	a_1
ν_5	121	2	a	ν_5	115	3	a"	ν_5	123	3	a_2
ν_6	156	0	b	ν_6	163	1	a"	ν_6	182	0	b_2
ν_7	221	1	b	ν_7	210	1	a'	ν_7	206	0	b_1
ν_8	229	2	a	ν_8	231	1	a'	ν_8	233	1	a_1
ν_9	299	12	a	ν_9	314	2	a"	ν_9	318	2	a_2
ν_{10}	371	0	a	ν_{10}	345	3	a'	ν_{10}	357	8	a_1
ν_{11}	376	1	b	ν_{11}	383	0	a"	ν_{11}	385	1	b_2
ν_{12}	410	11	a	ν_{12}	411	9	a'	ν_{12}	413	0	b_1
ν_{13}	411	1	b	ν_{13}	411	1	a"	ν_{13}	414	0	a_2
ν_{14}	422	1	b	ν_{14}	440	4	a"	ν_{14}	447	4	b_2
ν_{15}	482	1	b	ν_{15}	467	5	a'	ν_{15}	460	0	b_1
ν_{16}	512	11	a	ν_{16}	518	0	a"	ν_{16}	519	0	a_2
ν_{17}	578	0	b	ν_{17}	578	0	a'	ν_{17}	590	0	b_1
ν_{18}	603	8	a	ν_{18}	604	2	a"	ν_{18}	619	2	a_2
ν_{19}	621	14	a	ν_{19}	621	15	a'	ν_{19}	621	18	a_1
ν_{20}	627	8	b	ν_{20}	627	8	a"	ν_{20}	627	9	b_2
ν_{21}	629	2	a	ν_{21}	628	1	a'	ν_{21}	629	1	a_1
ν_{22}	691	1	b	ν_{22}	691	0	a"	ν_{22}	692	1	b_2
ν_{23}	700	0	b	ν_{23}	700	8	a'	ν_{23}	705	0	b_1
ν_{24}	700	3	a	ν_{24}	702	0	a"	ν_{24}	705	1	a_2
ν_{25}	706	41	a	ν_{25}	706	40	a'	ν_{25}	707	46	a_1
ν_{26}	743	1	b	ν_{26}	743	1	a"	ν_{26}	743	1	b_2
ν_{27}	767	0	b	ν_{27}	767	5	a'	ν_{27}	778	0	b_1
ν_{28}	772	7	a	ν_{28}	772	7	a"	ν_{28}	783	7	a_2
ν_{29}	810	0	b	ν_{29}	807	27	a'	ν_{29}	829	0	b_1
ν_{30}	838	22	a	ν_{30}	839	0	a"	ν_{30}	846	1	b_1
ν_{31}	839	1	b	ν_{31}	840	18	a'	ν_{31}	846	0	a_2
ν_{32}	892	5	a	ν_{32}	889	4	a"	ν_{32}	907	4	a_2
ν_{33}	916	0	a	ν_{33}	917	1	a'	ν_{33}	933	0	b_1

Continued on next page

C_2				C_S				C_{2v}			
Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry
ν_{34}	918	0	b	ν_{34}	917	0	a"	ν_{34}	934	0	a_2
ν_{35}	947	5	a	ν_{35}	947	6	a’	ν_{35}	947	7	a_1
ν_{36}	977	4	a	ν_{36}	977	0	a"	ν_{36}	979	0	b_2
ν_{37}	977	3	b	ν_{37}	977	8	a’	ν_{37}	993	0	b_1
ν_{38}	979	1	b	ν_{38}	979	0	a"	ν_{38}	993	0	a_2
ν_{39}	997	1	a	ν_{39}	998	0	a’	ν_{39}	1006	499	a_1
ν_{40}	998	1	b	ν_{40}	998	2	a"	ν_{40}	1007	5	b_2
ν_{41}	1006	388	a	ν_{41}	1006	384	a’	ν_{41}	1011	2	a_2
ν_{42}	1007	7	b	ν_{42}	1007	4	a"	ν_{42}	1011	0	b_1
ν_{43}	1048	163	a	ν_{43}	1047	158	a’	ν_{43}	1048	193	a_1
ν_{44}	1052	2	b	ν_{44}	1051	1	a"	ν_{44}	1052	2	b_2
ν_{45}	1085	295	a	ν_{45}	1083	298	a’	ν_{45}	1091	345	a_1
ν_{46}	1101	0	b	ν_{46}	1100	0	a"	ν_{46}	1103	0	b_2
ν_{47}	1103	14	a	ν_{47}	1102	9	a’	ν_{47}	1106	63	a_1
ν_{48}	1172	17	b	ν_{48}	1172	20	a"	ν_{48}	1172	23	b_2
ν_{49}	1172	5	a	ν_{49}	1172	2	a’	ν_{49}	1172	2	a_1
ν_{50}	1199	2	b	ν_{50}	1198	0	a"	ν_{50}	1202	0	b_2
ν_{51}	1201	238	a	ν_{51}	1199	230	a’	ν_{51}	1204	357	a_1
ν_{52}	1217	0	b	ν_{52}	1215	1	a"	ν_{52}	1220	0	b_2
ν_{53}	1244	95	a	ν_{53}	1243	103	a’	ν_{53}	1245	112	a_1
ν_{54}	1291	55	b	ν_{54}	1290	51	a"	ν_{54}	1293	65	b_2
ν_{55}	1313	220	a	ν_{55}	1313	209	a’	ν_{55}	1313	325	a_1
ν_{56}	1325	5	b	ν_{56}	1324	4	a"	ν_{56}	1328	7	b_2
ν_{57}	1353	8	a	ν_{57}	1352	6	a’	ν_{57}	1355	21	a_1
ν_{58}	1354	12	b	ν_{58}	1353	16	a"	ν_{58}	1356	16	b_2
ν_{59}	1371	358	a	ν_{59}	1370	319	a’	ν_{59}	1374	544	a_1
ν_{60}	1462	10	b	ν_{60}	1462	14	a"	ν_{60}	1463	14	b_2
ν_{61}	1466	433	a	ν_{61}	1466	408	a’	ν_{61}	1467	748	a_1
ν_{62}	1493	3035	a	ν_{62}	1492	2842	a’	ν_{62}	1492	4296	a_1
ν_{63}	1510	0	b	ν_{63}	1509	0	a"	ν_{63}	1512	1	b_2
ν_{64}	1530	4380	a	ν_{64}	1530	4452	a’	ν_{64}	1530	4638	a_1
ν_{65}	1582	85	b	ν_{65}	1581	80	a"	ν_{65}	1582	101	b_2
ν_{66}	1614	25	a	ν_{66}	1613	1	a"	ν_{66}	1615	3	b_2
ν_{67}	1614	2	b	ν_{67}	1613	25	a’	ν_{67}	1615	30	a_1
ν_{68}	1642	2438	a	ν_{68}	1642	2484	a’	ν_{68}	1641	2863	a_1

Continued on next page

C_2				C_S				C_{2v}			
Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity (Å^4/amu)	Symmetry
ν_{69}	1642	137	b	ν_{69}	1642	123	a"	ν_{69}	1642	143	b_2
ν_{70}	3170	23	a	ν_{70}	3170	41	a'	ν_{70}	3170	5	b_2
ν_{71}	3170	23	b	ν_{71}	3170	5	a"	ν_{71}	3170	48	a_1
ν_{72}	3176	189	b	ν_{72}	3176	220	a"	ν_{72}	3177	229	b_2
ν_{73}	3176	61	a	ν_{73}	3176	26	a'	ν_{73}	3177	26	a_1
ν_{74}	3186	23	b	ν_{74}	3186	2	a"	ν_{74}	3187	11	b_2
ν_{75}	3186	147	a	ν_{75}	3186	167	a'	ν_{75}	3187	159	a_1
ν_{76}	3193	42	b	ν_{76}	3193	51	a"	ν_{76}	3195	38	b_2
ν_{77}	3193	44	a	ν_{77}	3193	30	a'	ν_{77}	3196	165	a_1
ν_{78}	3200	50	b	ν_{78}	3200	16	a"	ν_{78}	3201	6	b_2
ν_{79}	3200	679	a	ν_{79}	3200	725	a'	ν_{79}	3202	587	a_1
ν_{80}	3206	63	b	ν_{80}	3205	62	a"	ν_{80}	3211	62	b_2
ν_{81}	3220	160	a	ν_{81}	3219	159	a'	ν_{81}	3224	162	a_1

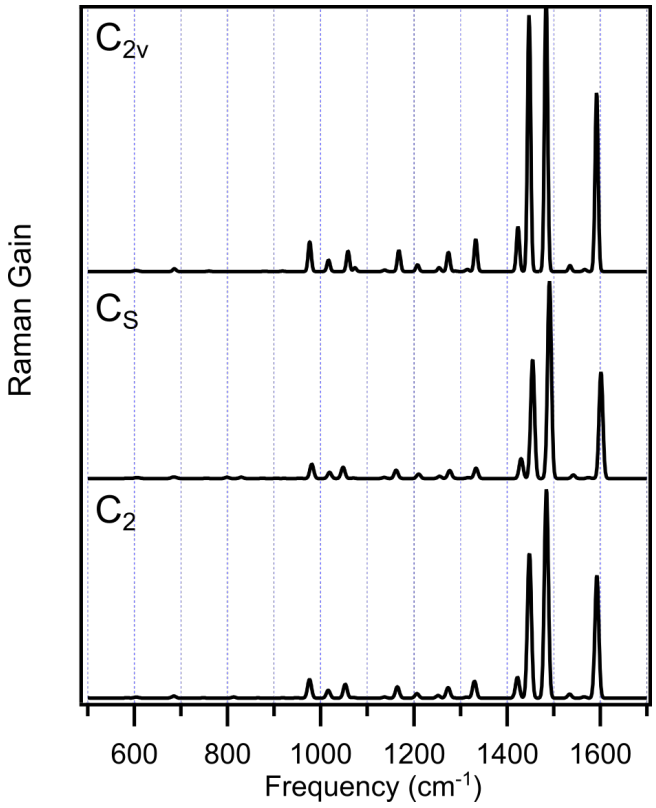


Figure S1: Calculated ground-state (S_0) vibrational spectra for the three conformations of DPT (B3LYP/aug-cc-pVDZ). Values from Table S2.

Table S3: Optimized excited-state (S_1) geometry of DPT.

Atom	B3LYP/aug-cc-pVDZ Cartesian Coordinates (Å)			EOM-CCSD/6-31G* Cartesian Coordinates (Å)		
	X	Y	Z	X	Y	Z
S	0.000000	0.000000	0.764300	0.000000	0.000000	0.758211
C	0.000000	1.275268	-0.478907	0.000000	1.266118	-0.469087
C	0.000000	0.690854	-1.781879	0.000000	0.688140	-1.773531
C	0.000000	-0.690854	-1.781879	0.000000	-0.688140	-1.773531
C	0.000000	-1.275268	-0.478907	0.000000	-1.266118	-0.469087
C	0.000000	2.652956	-0.122033	0.000000	2.643094	-0.117140
C	0.000000	-2.652956	-0.122033	0.000000	-2.643094	-0.117140
C	0.000000	3.072524	1.247115	0.000000	3.065630	1.246631
C	0.000000	4.419278	1.581720	0.000000	4.413710	1.574020
C	0.000000	5.405765	0.581164	0.000000	5.395163	0.573900
C	0.000000	5.017537	-0.772088	0.000000	5.001212	-0.773968
C	0.000000	3.676940	-1.124223	0.000000	3.660113	-1.120787
C	0.000000	-3.676940	-1.124223	0.000000	-3.660113	-1.120787
C	0.000000	-5.017537	-0.772088	0.000000	-5.001212	-0.773968
C	0.000000	-5.405765	0.581164	0.000000	-5.395163	0.573900
C	0.000000	-4.419278	1.581720	0.000000	-4.413710	1.574020
C	0.000000	-3.072524	1.247115	0.000000	-3.065630	1.246631
H	0.000000	1.289069	-2.689415	0.000000	1.286242	-2.679247
H	0.000000	-1.289069	-2.689415	0.000000	-1.286242	-2.679247
H	0.000000	2.323008	2.038800	0.000000	2.323648	2.043965
H	0.000000	4.711991	2.632324	0.000000	4.708642	2.622764
H	0.000000	6.462152	0.849887	0.000000	6.451079	0.838402
H	0.000000	5.778340	-1.553469	0.000000	5.756160	-1.559325
H	0.000000	3.402271	-2.177743	0.000000	3.382141	-2.172465
H	0.000000	-3.402271	-2.177743	0.000000	-3.382141	-2.172465
H	0.000000	-5.778340	-1.553469	0.000000	-5.756160	-1.559325
H	0.000000	-6.462152	0.849887	0.000000	-6.451079	0.838402
H	0.000000	-4.711991	2.632324	0.000000	-4.708642	2.622764
H	0.000000	-2.323008	2.038800	0.000000	-2.323648	2.043965

Table S4: Calculated excited-state (S_1) vibrational frequencies and off-resonance Raman activities of DPT. Raman activities from EOM-CCSD are limited to a subset of a_1-symmetry modes due to computational cost.

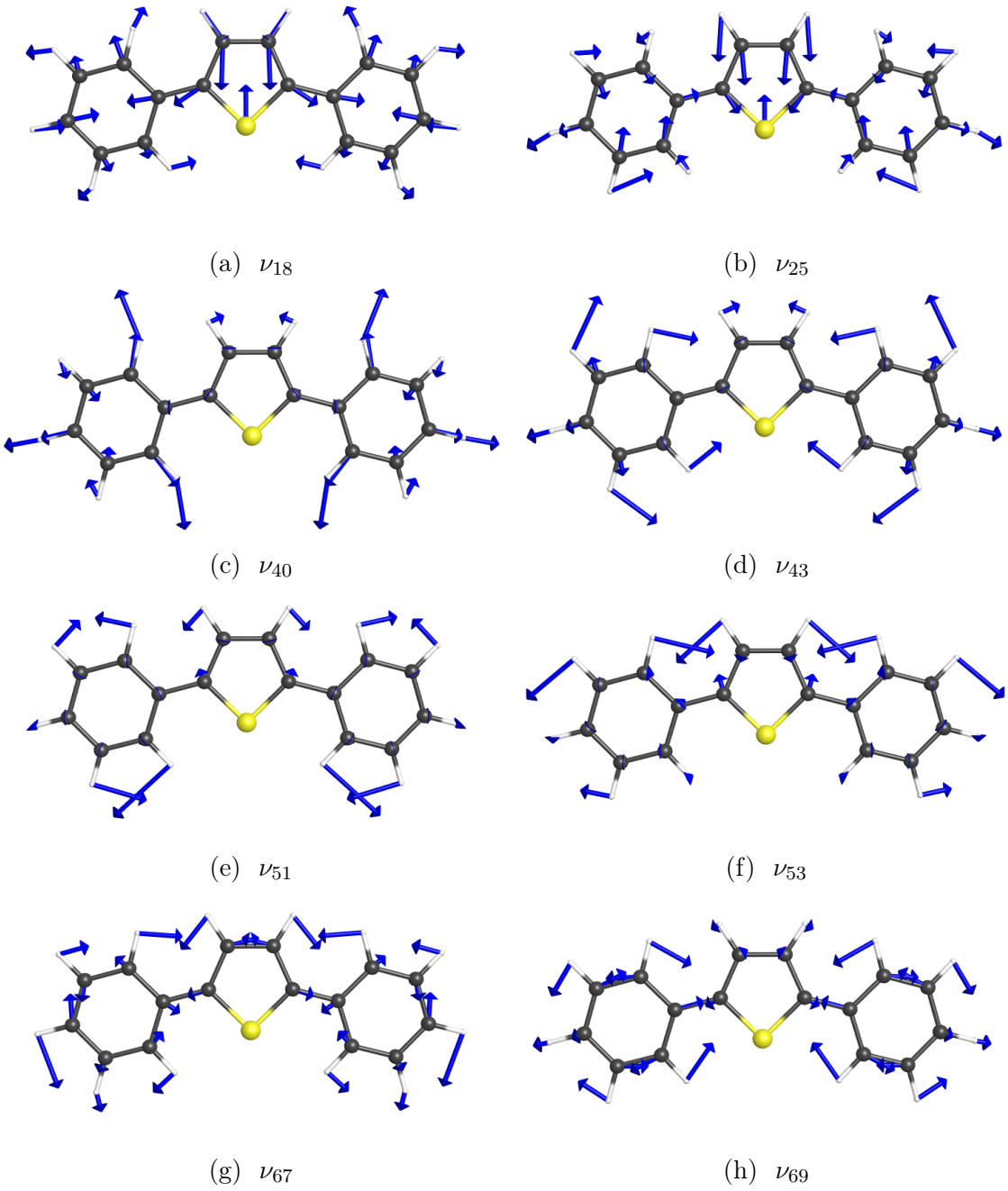
B3LYP/aug-cc-pVDZ				EOM-CCSD/6-31G*			
Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry
ν_1	36	0	b_1	ν_1	27		b_1
ν_2	55	0	a_2	ν_2	31		a_2
ν_3	76	6	a_1	ν_3	39		b_1
ν_4	93	1	b_1	ν_4	82		a_1
ν_5	107	1	a_2	ν_5	90		a_2
ν_6	165	1	b_1	ν_6	137		b_1
ν_7	174	3	b_2	ν_7	185		b_2
ν_8	229	855	a_1	ν_8	237		a_1
ν_9	281	1	a_2	ν_9	240		a_2
ν_{10}	361	304	a_1	ν_{10}	363		a_1
ν_{11}	382	3	b_2	ν_{11}	373		b_1
ν_{12}	384	2	b_1	ν_{12}	387		b_2
ν_{13}	415	0	a_2	ν_{13}	389		a_2
ν_{14}	416	0	b_1	ν_{14}	390		b_1
ν_{15}	440	12	b_2	ν_{15}	441		a_2
ν_{16}	475	27	a_2	ν_{16}	448		b_2
ν_{17}	541	0	b_1	ν_{17}	483		b_1
ν_{18}	590	3360	a_1	ν_{18}	495		a_2
ν_{19}	613	0	b_2	ν_{19}	515		b_1
ν_{20}	616	17	a_2	ν_{20}	519		a_2
ν_{21}	618	9	a_1	ν_{21}	606	460	a_1
ν_{22}	651	16	b_2	ν_{22}	615		b_2
ν_{23}	670	26	a_2	ν_{23}	625	50	a_1
ν_{24}	673	0	b_1	ν_{24}	656		b_2
ν_{25}	691	1407	a_1	ν_{25}	667		b_1
ν_{26}	697	0	b_2	ν_{26}	671		a_2
ν_{27}	735	1	b_1	ν_{27}	696		b_1
ν_{28}	779	37	a_2	ν_{28}	703		b_2
ν_{29}	785	0	b_1	ν_{29}	711	253	a_1
ν_{30}	809	1	a_2	ν_{30}	750		a_2
ν_{31}	811	2	b_1	ν_{31}	762		b_1
ν_{32}	884	17	a_2	ν_{32}	788		a_2

Continued on next page

B3LYP/aug-cc-pVDZ				EOM-CCSD/6-31G*			
Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry
ν_{33}	894	1	b_1	ν_{33}	818		b_1
ν_{34}	899	37	a_2	ν_{34}	822		a_2
ν_{35}	923	678	a_1	ν_{35}	894		a_2
ν_{36}	947	30	b_2	ν_{36}	894		b_1
ν_{37}	982	1	a_2	ν_{37}	903		b_1
ν_{38}	982	1	b_1	ν_{38}	904		a_2
ν_{39}	985	0	b_2	ν_{39}	950		a_1
ν_{40}	992	196	a_1	ν_{40}	971		b_2
ν_{41}	997	16	a_2	ν_{41}	996		b_2
ν_{42}	997	0	b_1	ν_{42}	1010		a_1
ν_{43}	1031	2981	a_1	ν_{43}	1062		a_1
ν_{44}	1032	75	b_2	ν_{44}	1065		b_2
ν_{45}	1080	337	b_2	ν_{45}	1112		b_2
ν_{46}	1100	1125	a_1	ν_{46}	1131		a_1
ν_{47}	1111	3250	a_1	ν_{47}	1152		a_1
ν_{48}	1136	610	b_2	ν_{48}	1171		b_2
ν_{49}	1167	35	a_1	ν_{49}	1209		a_1
ν_{50}	1172	30	b_2	ν_{50}	1214		b_2
ν_{51}	1198	5030	a_1	ν_{51}	1241		b_2
ν_{52}	1199	13	b_2	ν_{52}	1242		a_1
ν_{53}	1214	9709	a_1	ν_{53}	1260		a_1
ν_{54}	1315	0	b_2	ν_{54}	1340		b_2
ν_{55}	1319	466	a_1	ν_{55}	1348		a_1
ν_{56}	1327	0	b_2	ν_{56}	1356		b_2
ν_{57}	1367	9	a_1	ν_{57}	1389		a_1
ν_{58}	1367	380	b_2	ν_{58}	1390		b_2
ν_{59}	1416	5483	a_1	ν_{59}	1459		a_1
ν_{60}	1428	1190	b_2	ν_{60}	1478		b_2
ν_{61}	1461	5920	a_1	ν_{61}	1511		a_1
ν_{62}	1463	198	b_2	ν_{62}	1513		b_2
ν_{63}	1500	440	b_2	ν_{63}	1544		b_2
ν_{64}	1500	4941	a_1	ν_{64}	1559	5220	a_1
ν_{65}	1548	118864	a_1	ν_{65}	1620		b_2
ν_{66}	1548	146	b_2	ν_{66}	1621	4456	a_1
ν_{67}	1555	65556	a_1	ν_{67}	1632	22265	a_1

Continued on next page

B3LYP/aug-cc-pVDZ				EOM-CCSD/6-31G*			
Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry	Mode	Calc. Frequency (cm^{-1})	Raman Activity ($Å^4$/amu)	Symmetry
ν_{68}	1613	141	b_2	ν_{68}	1663		b_2
ν_{69}	1643	16075	a_1	ν_{69}	1705	5301	a_1
ν_{70}	3171	1	b_2	ν_{70}	3191		b_2
ν_{71}	3171	67	a_1	ν_{71}	3191		a_1
ν_{72}	3176	423	b_2	ν_{72}	3196		b_2
ν_{73}	3176	159	a_1	ν_{73}	3197		a_1
ν_{74}	3187	75	b_2	ν_{74}	3208		b_2
ν_{75}	3187	145	a_1	ν_{75}	3209		a_1
ν_{76}	3197	203	b_2	ν_{76}	3218		b_2
ν_{77}	3197	336	a_1	ν_{77}	3219		a_1
ν_{78}	3203	13	b_2	ν_{78}	3225		b_2
ν_{79}	3203	713	a_1	ν_{79}	3226		a_1
ν_{80}	3219	11	b_2	ν_{80}	3247		b_2
ν_{81}	3236	131	a_1	ν_{81}	3265		a_1



(a) ν_{18} (b) ν_{25}

(c) ν_{40} (d) ν_{43}

(e) ν_{51} (f) ν_{53}

(g) ν_{67} (h) ν_{69}

Figure S2: Displacement vectors for key vibrational modes in the S_1 excited state of DPT (B3LYP/aug-cc-pVDZ). Frequencies and Raman activities are in Table S4.

Table S5: Calculated excited-state (S_n) energy gradients along the S_1 normal mode coordinates of DPT for $n = 1$ to 10 using B3LYP/aug-cc-pVDZ. (See below for $n = 11$ to 20) Gradient values are scaled by 10^3, and oscillator strengths are included for each $S_n \leftarrow S_1$ transition. (All values calculated with B3LYP/aug-cc-pVDZ.)

		Electronic State Transition ($S_n \leftarrow S_1$)									
		Osc.Str. :	0	0.0389	0.0021	0.1088	0.0057	0.0000	0.0066	0.0001	0.0000	0.0000
Freq	Mode	Symmetry	1	2	3	4	5	6	7	8	9	10
36.1	ν_1	b_1	0.000	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003
55.4	ν_2	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
76.3	ν_3	a_1	0.000	-1.094	-0.020	1.053	1.053	-1.094	-0.020	-0.020	-0.020	-1.094
92.9	ν_4	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
106.7	ν_5	a_2	0.000	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
165.3	ν_6	b_1	0.000	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
173.8	ν_7	b_2	0.000	0.024	0.024	0.024	0.024	0.024	0.024	0.024	0.024	0.024
229.3	ν_8	a_1	0.000	0.313	-0.649	-1.610	-3.533	2.236	-2.572	-2.572	1.274	2.236
281.2	ν_9	a_2	0.000	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023
361.2	ν_{10}	a_1	-0.005	1.436	1.436	1.436	2.425	1.436	0.447	1.436	-1.531	-6.476
381.9	ν_{11}	b_2	0.000	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007
383.8	ν_{12}	b_1	0.000	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
414.9	ν_{13}	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
415.9	ν_{14}	b_1	0.000	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
440.3	ν_{15}	b_2	0.000	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024
475.1	ν_{16}	a_2	0.000	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027
541.5	ν_{17}	b_1	0.000	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
590.1	ν_{18}	a_1	0.000	5.854	5.032	2.565	7.498	4.210	-1.546	-3.190	1.743	-12.235
612.5	ν_{19}	b_2	0.000	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013
616.1	ν_{20}	a_2	0.000	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011
617.6	ν_{21}	a_1	0.005	1.403	0.427	2.378	-0.548	0.427	-5.425	0.427	0.427	-0.548
650.9	ν_{22}	b_2	0.000	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019
670.1	ν_{23}	a_2	0.000	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017
672.8	ν_{24}	b_1	0.000	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022
690.6	ν_{25}	a_1	0.002	3.677	4.587	1.857	0.946	3.677	8.228	8.228	0.946	-12.707
696.8	ν_{26}	b_2	0.000	-0.114	-0.114	-0.114	-0.114	-0.114	-0.114	-0.114	-0.114	0.837
735.0	ν_{27}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
778.7	ν_{28}	a_2	0.000	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004
784.6	ν_{29}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
808.6	ν_{30}	a_2	0.000	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
810.6	ν_{31}	b_1	0.000	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
884.1	ν_{32}	a_2	0.000	0.017	0.017	0.017	0.017	0.017	0.017	0.017	0.017	0.017
893.8	ν_{33}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Continued on next page

		Electronic State Transition ($S_n \leftarrow S_1$)									
		Osc.Str. :	0	0.0389	0.0021	0.1088	0.0057	0.0000	0.0066	0.0001	0.0000	0.0000
Freq	Mode	Symmetry	1	2	3	4	5	6	7	8	9	10
899.0	ν_{34}	a_2	0.000	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005
923.0	ν_{35}	a_1	0.003	-3.908	-5.024	-1.675	10.605	0.558	-2.792	-0.559	1.674	7.256
946.9	ν_{36}	b_2	0.000	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041
982.0	ν_{37}	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
982.3	ν_{38}	b_1	0.000	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011
984.5	ν_{39}	b_2	0.000	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050
992.0	ν_{40}	a_1	-0.009	-1.688	-0.632	2.535	1.479	-5.911	1.479	-0.632	-6.967	-3.800
996.5	ν_{41}	a_2	0.000	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012
996.6	ν_{42}	b_1	0.000	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
1031.2	ν_{43}	a_1	0.000	3.236	4.875	6.514	3.236	-3.321	4.875	4.875	-1.682	-0.042
1032.5	ν_{44}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1080.4	ν_{45}	b_2	0.000	0.118	0.118	0.118	0.118	0.118	0.118	0.118	0.118	0.118
1100.4	ν_{46}	a_1	-0.005	1.470	1.470	1.470	3.389	-0.449	3.389	1.470	-0.449	5.308
1111.4	ν_{47}	a_1	-0.006	6.965	-1.141	-1.141	2.912	0.885	-1.141	-3.168	-1.141	-3.168
1136.3	ν_{48}	b_2	0.000	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087
1166.5	ν_{49}	a_1	0.006	5.455	0.768	-8.605	-6.262	0.768	3.112	-1.575	-1.575	-1.575
1172.2	ν_{50}	b_2	0.000	0.051	0.051	0.051	0.051	0.051	0.051	0.051	0.051	0.051
1197.6	ν_{51}	a_1	0.006	5.428	7.533	-7.204	-9.309	1.217	3.323	7.533	1.217	-2.993
1199.0	ν_{52}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1213.5	ν_{53}	a_1	-0.010	8.045	13.820	9.970	19.596	-1.582	9.970	13.820	2.269	8.045
1315.4	ν_{54}	b_2	0.000	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020
1319.0	ν_{55}	a_1	0.000	-8.647	-3.407	5.325	-5.154	1.832	-10.393	-3.407	0.086	-1.661
1326.8	ν_{56}	b_2	0.000	0.043	0.043	0.043	0.043	0.043	0.043	0.043	0.043	0.043
1366.5	ν_{57}	a_1	-0.008	-1.136	0.390	1.917	-2.662	1.917	-4.189	-1.136	3.443	4.969
1366.6	ν_{58}	b_2	-0.004	-1.273	0.229	0.229	-1.273	1.731	-2.776	0.229	0.229	1.731
1416.0	ν_{59}	a_1	-0.008	-7.718	-0.815	3.327	-2.196	6.088	-0.815	-0.815	6.088	8.849
1428.5	ν_{60}	b_2	0.000	1.572	0.033	0.033	0.033	0.033	0.033	0.033	0.033	0.033
1460.9	ν_{61}	a_1	0.000	-11.149	-0.926	6.376	1.995	-2.387	-9.689	-0.926	-3.847	-8.228
1462.8	ν_{62}	b_2	0.000	-0.223	-0.223	-0.223	-0.223	1.217	-0.223	-0.223	-0.223	-0.223
1499.6	ν_{63}	b_2	-0.004	-2.087	-0.542	-0.542	-0.542	1.003	-0.542	-0.542	-0.542	1.003
1500.4	ν_{64}	a_1	0.016	7.851	0.718	2.145	9.278	-3.562	6.425	2.145	-2.135	2.145
1548.3	ν_{65}	a_1	0.007	11.091	1.195	-2.516	3.669	7.380	11.091	1.195	6.143	7.380
1548.8	ν_{66}	b_2	0.010	15.681	2.438	-5.990	2.438	8.457	15.681	1.234	9.661	10.865
1555.8	ν_{67}	a_1	0.003	-1.944	-0.809	23.032	18.491	9.409	-3.079	0.326	12.815	18.491
1613.4	ν_{68}	b_2	0.000	1.097	0.032	0.032	0.032	0.032	0.032	0.032	0.032	0.032
1644.2	ν_{69}	a_1	0.006	26.937	43.126	6.701	21.878	18.842	21.878	38.067	16.819	16.819
3170.7	ν_{70}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Continued on next page

			Electronic State Transition ($S_n \leftarrow S_1$)									
		Osc.Str. :	0	0.0389	0.0021	0.1088	0.0057	0.0000	0.0066	0.0001	0.0000	0.0000
Freq	Mode	Symmetry	1	2	3	4	5	6	7	8	9	10
3170.7	ν_{71}	a_1	0.000	3.050	3.050	-1.714	0.668	3.050	0.668	0.668	0.668	-1.714
3176.1	ν_{72}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
3176.2	ν_{73}	a_1	0.006	2.703	0.324	-2.055	0.324	2.703	0.324	0.324	-2.055	-2.055
3186.9	ν_{74}	b_2	0.000	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
3187.2	ν_{75}	a_1	0.000	1.843	4.214	1.843	-0.529	-2.901	-0.529	1.843	-0.529	1.843
3196.5	ν_{76}	b_2	0.000	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
3196.9	ν_{77}	a_1	-0.006	-0.484	-0.484	1.887	-0.484	1.887	1.887	1.887	-7.596	1.887
3202.6	ν_{78}	b_2	0.000	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
3203.4	ν_{79}	a_1	-0.006	-5.919	-5.919	-3.551	-1.183	3.554	-3.551	-3.551	8.291	3.554
3219.3	ν_{80}	b_2	0.000	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
3235.7	ν_{81}	a_1	0.013	-4.783	-0.048	-0.048	-2.416	9.421	2.319	2.319	-2.416	14.156

Table S6: Calculated excited-state (S_n) energy gradients along the S_1 normal mode coordinates of DPT for $n = 11$ to 20 using B3LYP/aug-cc-pVDZ. (See above for $n = 1$ to 10) Gradient values are scaled by 10^3, and oscillator strengths are included for each $S_n \leftarrow S_1$ transition. (All values calculated with B3LYP/aug-cc-pVDZ.)

		Electronic State Transition ($S_n \leftarrow S_1$)										
		Osc.Str. :	0.0002	0.0006	0.0068	0.0000	0.0049	0.0020	0.0000	0.0008	0.0005	0.0000
Freq	Mode	Symmetry	11	12	13	14	15	16	17	18	19	20
36.1	ν_1	b_1	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003
55.4	ν_2	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
76.3	ν_3	a_1	-0.020	-1.094	-0.020	-1.094	-0.020	-0.020	-0.020	-1.094	1.053	-0.020
92.9	ν_4	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
106.7	ν_5	a_2	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
165.3	ν_6	b_1	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
173.8	ν_7	b_2	0.024	0.024	0.024	0.024	0.024	0.024	0.024	0.024	0.024	0.024
229.3	ν_8	a_1	1.274	-3.533	1.274	0.313	-1.610	2.236	1.274	0.313	-2.572	-2.572
281.2	ν_9	a_2	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023	-0.023
361.2	ν_{10}	a_1	1.436	2.425	0.447	-0.542	1.436	1.436	0.447	1.436	3.414	1.436
381.9	ν_{11}	b_2	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007	-0.007
383.8	ν_{12}	b_1	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
414.9	ν_{13}	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
415.9	ν_{14}	b_1	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
440.3	ν_{15}	b_2	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024	-0.024
475.1	ν_{16}	a_2	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027	0.027
541.5	ν_{17}	b_1	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
590.1	ν_{18}	a_1	5.854	7.498	5.854	0.921	0.099	4.210	6.676	5.854	7.498	3.387
612.5	ν_{19}	b_2	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013	-0.013
616.1	ν_{20}	a_2	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011
617.6	ν_{21}	a_1	0.427	1.403	0.427	-0.548	-0.548	0.427	0.427	-0.548	5.304	-0.548
650.9	ν_{22}	b_2	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019	-0.019
670.1	ν_{23}	a_2	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017	-0.017
672.8	ν_{24}	b_1	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022	-0.022
690.6	ν_{25}	a_1	3.677	3.677	4.587	-1.784	-0.874	3.677	4.587	3.677	3.677	2.767
696.8	ν_{26}	b_2	-0.114	-0.114	-0.114	0.837	-0.114	-0.114	-0.114	0.837	-0.114	-0.114
735.0	ν_{27}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
778.7	ν_{28}	a_2	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004	-0.004
784.6	ν_{29}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
808.6	ν_{30}	a_2	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
810.6	ν_{31}	b_1	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
884.1	ν_{32}	a_2	0.017	0.017	0.017	0.017	0.017	0.017	0.017	0.017	0.017	0.017
893.8	ν_{33}	b_1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Continued on next page

		Electronic State Transition ($S_n \leftarrow S_1$)										
		Osc.Str. :	0.0002	0.0006	0.0068	0.0000	0.0049	0.0020	0.0000	0.0008	0.0005	0.0000
Freq	Mode	Symmetry	11	12	13	14	15	16	17	18	19	20
899.0	ν_{34}	a_2	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005
923.0	ν_{35}	a_1	-0.559	2.790	-0.559	1.674	-6.141	0.558	-0.559	0.558	-0.559	-1.675
946.9	ν_{36}	b_2	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041	-0.041
982.0	ν_{37}	a_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
982.3	ν_{38}	b_1	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011
984.5	ν_{39}	b_2	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050	-0.050
992.0	ν_{40}	a_1	-5.911	0.424	-6.967	-5.911	-3.800	-5.911	-4.856	-5.911	2.535	-0.632
996.5	ν_{41}	a_2	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012	-0.012
996.6	ν_{42}	b_1	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
1031.2	ν_{43}	a_1	-3.321	3.236	-3.321	-0.042	9.792	-0.042	-1.682	-0.042	8.153	8.153
1032.5	ν_{44}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1080.4	ν_{45}	b_2	0.118	0.118	0.118	0.118	0.118	0.118	0.118	-1.618	0.118	0.118
1100.4	ν_{46}	a_1	-2.368	1.470	-0.449	1.470	-2.368	-2.368	-0.449	1.470	3.389	1.470
1111.4	ν_{47}	a_1	-1.141	4.938	0.885	0.885	4.938	-3.168	2.912	0.885	4.938	4.938
1136.3	ν_{48}	b_2	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087	-0.087
1166.5	ν_{49}	a_1	0.768	-3.918	3.112	-3.918	0.768	0.768	7.798	-3.918	-1.575	0.768
1172.2	ν_{50}	b_2	0.051	0.051	0.051	0.051	0.051	0.051	0.051	0.051	0.051	0.051
1197.6	ν_{51}	a_1	1.217	-7.204	3.323	1.217	3.323	3.323	-0.888	1.217	-7.204	-0.888
1199.0	ν_{52}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1213.5	ν_{53}	a_1	-3.507	11.895	-1.582	0.343	-5.432	-1.582	2.269	0.343	21.522	6.119
1315.4	ν_{54}	b_2	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020	-0.020
1319.0	ν_{55}	a_1	5.325	1.832	3.578	1.832	7.071	5.325	3.578	3.578	-8.647	0.086
1326.8	ν_{56}	b_2	0.043	0.043	0.043	0.043	0.043	0.043	0.043	0.043	0.043	0.043
1366.5	ν_{57}	a_1	1.917	1.917	0.390	1.917	3.443	1.917	0.390	1.917	-4.189	0.390
1366.6	ν_{58}	b_2	0.229	1.731	0.229	1.731	1.731	1.731	0.229	1.731	-1.273	0.229
1416.0	ν_{59}	a_1	6.088	-4.957	4.707	6.088	11.610	6.088	4.707	3.327	3.327	7.469
1428.5	ν_{60}	b_2	0.033	0.033	0.033	0.033	0.033	0.033	0.033	0.033	0.033	0.033
1460.9	ν_{61}	a_1	-2.387	-0.926	-0.926	-2.387	-2.387	-2.387	-0.926	-2.387	-11.149	-3.847
1462.8	ν_{62}	b_2	-0.223	-0.223	-0.223	1.217	-0.223	-0.223	-0.223	1.217	-0.223	-0.223
1499.6	ν_{63}	b_2	1.003	-2.087	-0.542	1.003	1.003	1.003	-0.542	1.003	-2.087	-0.542
1500.4	ν_{64}	a_1	-3.562	10.705	-4.988	-0.708	-13.548	-2.135	-4.988	-3.562	10.705	-2.135
1548.3	ν_{65}	a_1	7.380	7.380	6.143	7.380	11.091	6.143	6.143	7.380	17.276	14.802
1548.8	ν_{66}	b_2	9.661	8.457	9.661	10.865	15.681	8.457	9.661	8.457	22.905	20.497
1555.8	ν_{67}	a_1	9.409	27.574	10.544	11.679	18.491	9.409	10.544	9.409	27.574	21.897
1613.4	ν_{68}	b_2	0.032	0.032	0.032	0.032	0.032	0.032	0.032	0.032	0.032	0.032
1644.2	ν_{69}	a_1	18.842	15.807	17.831	18.842	8.724	18.842	17.831	16.819	28.961	17.831
3170.7	ν_{70}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Continued on next page

		Electronic State Transition ($S_n \leftarrow S_1$)										
		Osc.Str. :	0.0002	0.0006	0.0068	0.0000	0.0049	0.0020	0.0000	0.0008	0.0005	0.0000
Freq	Mode	Symmetry	11	12	13	14	15	16	17	18	19	20
3170.7	ν_{71}	a_1	3.050	-1.714	0.668	3.050	0.668	3.050	5.431	-1.714	3.050	0.668
3176.1	ν_{72}	b_2	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
3176.2	ν_{73}	a_1	-2.055	-2.055	5.081	-2.055	0.324	2.703	0.324	2.703	2.703	0.324
3186.9	ν_{74}	b_2	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
3187.2	ν_{75}	a_1	-2.901	-0.529	4.214	-0.529	-0.529	-5.273	-0.529	1.843	1.843	-0.529
3196.5	ν_{76}	b_2	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
3196.9	ν_{77}	a_1	-5.225	1.887	-2.854	1.887	4.257	-2.854	-0.484	-2.854	1.887	1.887
3202.6	ν_{78}	b_2	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006	-0.006
3203.4	ν_{79}	a_1	1.186	-1.183	5.922	-1.183	-3.551	1.186	1.186	3.554	-5.919	-3.551
3219.3	ν_{80}	b_2	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
3235.7	ν_{81}	a_1	7.054	-0.048	2.319	-0.048	-0.048	16.523	2.319	-0.048	-2.416	-0.048

Table S7: Calculated excited-state (S_n) energy gradients along the S_1 normal mode coordinates of DPT for $n = 1$ to 14 using EOM-CCSD/6-31G*. Gradient values are scaled by 10^3, and oscillator strengths are included for each $S_n \leftarrow S_1$ transition. (All values calculated with EOM-CCSD/6-31G*.)

			Electronic State Transition ($S_n \leftarrow S_1$)						
		Osc.Str. :	0	0.0135	0.0021	0.0747	0.4639	0.0000	0.0099
Freq	Mode	Symmetry	1	2	3	4	5	6	7
606.4	ν_{21}	a_1	0.000	5.468	5.468	5.468	2.136	-22.020	7.134
624.8	ν_{23}	a_1	0.000	-0.239	-0.239	1.711	-1.214	1.711	0.736
711.2	ν_{29}	a_1	0.000	4.952	6.741	0.481	-4.885	-27.243	2.269
1558.9	ν_{64}	a_1	0.000	-8.758	-5.852	-11.664	-4.399	-0.040	-13.117
1620.9	ν_{66}	a_1	0.000	8.924	7.738	1.808	0.622	4.180	24.344
1631.9	ν_{67}	a_1	0.000	-20.355	-21.379	-26.499	-3.971	-26.499	-33.667
1705.3	ν_{69}	a_1	0.000	41.515	45.561	25.333	-14.110	28.367	16.231

			Electronic State Transition ($S_n \leftarrow S_1$)						
		Osc.Str. :	0.0001	0.0558	0.0190	0.0000	0.0004	0.0001	0.0000
Freq	Mode	Symmetry	8	9	10	11	12	13	14
606.4	ν_{21}	a_1	1.303	1.303	-0.363	-12.857	1.303	2.969	14.631
624.8	ν_{23}	a_1	-1.214	-0.239	1.711	3.660	-0.239	1.711	-2.188
711.2	ν_{29}	a_1	4.058	4.952	-5.779	-24.560	1.375	-14.723	8.530
1558.9	ν_{64}	a_1	-0.040	2.867	11.585	-21.835	5.773	2.867	-5.852
1620.9	ν_{66}	a_1	5.366	-14.798	6.552	17.227	11.297	-2.937	6.552
1631.9	ν_{67}	a_1	-0.899	-10.115	-15.235	-72.578	-36.739	-8.067	8.316
1705.3	ν_{69}	a_1	40.504	37.470	-6.019	55.674	41.515	18.254	-11.076

Table S8: Calculated excited-state (S_n) energy gradients of DPT for $n = 1$ to 20 using EOM-CCSD//TD-DFT. Optimized geometry and vibrational frequencies are from B3LYP/aug-cc-pVDZ, gradients and oscillator strengths are from EOM-CCSD/aug-cc-pVDZ.

		EOM-CCSD	Electronic State Transition ($S_n \leftarrow S_1$)									
TD-DFT		Osc.Str. :	0	0.0278	0.0039	0.0097	0.0638	0.3192	0.0002	0.0000	0.0000	0.1911
Freq	Mode	Symmetry	1	2	3	4	5	6	7	8	9	10
590.1	ν_{18}	a_1	-0.802	5.434	5.434	4.613	4.613	2.971	4.613	5.434	-15.091	3.792
617.6	ν_{21}	a_1	-0.302	1.054	1.054	0.080	-1.869	1.054	1.054	0.080	0.080	-1.869
690.6	ν_{25}	a_1	-2.089	2.056	3.881	1.143	-0.683	-4.334	0.000	1.143	-17.114	-1.596
1500.4	ν_{64}	a_1	1.272	9.070	6.368	-3.087	10.420	5.017	-4.438	-3.087	-0.386	17.174
1548.3	ν_{65}	a_1	0.000	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002
1555.8	ν_{67}	a_1	2.255	12.811	17.266	13.925	25.061	7.243	12.811	15.039	22.834	19.493
1644.2	ν_{69}	a_1	9.138	39.596	48.667	28.510	32.541	11.376	29.517	25.486	30.525	10.368

		EOM-CCSD	Electronic State Transition ($S_n \leftarrow S_1$)									
TD-DFT		Osc.Str. :	0.0053	0.0001	0.0000	0.0039	0.0000	0.0012	0.0124	0.0000	0.0119	0.0004
Freq	Mode	Symmetry	11	12	13	14	15	16	17	18	19	20
590.1	ν_{18}	a_1	-0.802	5.434	5.434	4.613	4.613	2.971	4.613	5.434	-15.091	3.792
617.6	ν_{21}	a_1	-0.302	1.054	1.054	0.080	-1.869	1.054	1.054	0.080	0.080	-1.869
690.6	ν_{25}	a_1	-2.089	2.056	3.881	1.143	-0.683	-4.334	0.000	1.143	-17.114	-1.596
1500.4	ν_{64}	a_1	1.272	9.070	6.368	-3.087	10.420	5.017	-4.438	-3.087	-0.386	17.174
1548.3	ν_{65}	a_1	0.000	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002
1555.8	ν_{67}	a_1	2.255	12.811	17.266	13.925	25.061	7.243	12.811	15.039	22.834	19.493
1644.2	ν_{69}	a_1	9.138	39.596	48.667	28.510	32.541	11.376	29.517	25.486	30.525	10.368

Table S9: Calculated excited-state (S_n) energy gradients of DPT for $n = 1$ to 10 using EOM-CCSD/aug-cc-pVDZ//EOM-CCSD/6-31G*. Optimized geometry and vibrational frequencies are from EOM-CCSD/6-31G*, gradients and oscillator strengths are from EOM-CCSD/aug-cc-pVDZ.

		aug-cc-pVDZ	Electronic State Transition ($S_n \leftarrow S_1$)									
6-31G*		Osc.Str. :	0	0.0267	0.0039	0.0100	0.0716	0.0002	0.2883	0.0000	0.0000	0.2173
Freq	Mode	Symmetry	1	2	3	4	5	6	7	8	9	10
606.4	ν_{21}	a_1	-1.324	4.737	4.737	3.071	3.904	19.731	3.071	46.276	73.581	3.071
624.8	ν_{23}	a_1	0.509	-1.737	-0.763	0.212	2.162	-0.763	-0.763	-21.850	-64.370	2.162
711.2	ν_{29}	a_1	-7.785	-4.332	-1.649	-4.332	-6.120	-8.803	-8.803	19.785	58.974	-7.015
1558.9	ν_{64}	a_1	-2.060	-9.944	-5.585	4.587	-9.944	-7.038	-7.038	-0.344	-0.054	-17.209
1620.9	ν_{66}	a_1	-0.278	10.296	6.738	6.738	0.807	-18.171	4.366	-39.031	-26.434	15.041
1631.9	ν_{67}	a_1	-2.711	-17.471	-19.519	-16.447	-27.711	-33.855	-12.351	-57.262	-90.910	-29.759
1705.3	ν_{69}	a_1	6.052	39.341	47.432	28.216	32.262	28.216	12.034	0.504	0.585	10.012

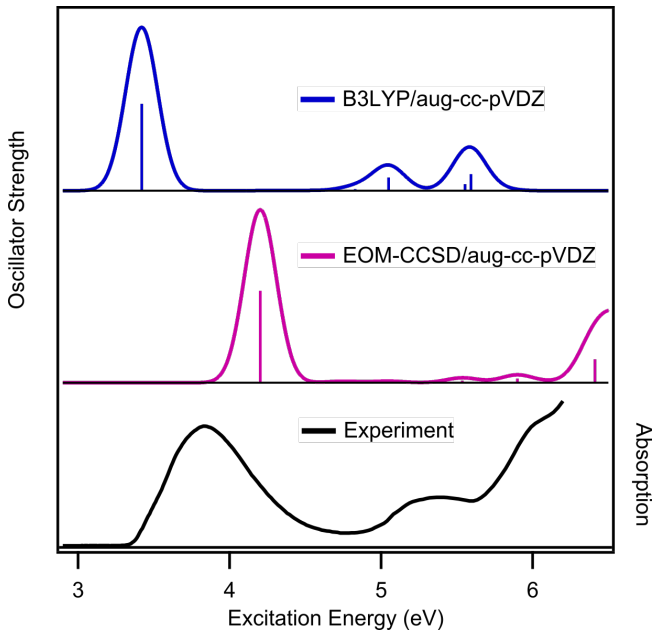


Figure S3: Comparison of calculated $S_n \leftarrow S_0$ transition energies and oscillator strengths for DPT (at C_{2v} geometry) with the experimental ground-state absorption spectrum. Calculated spectra include 0.25 eV Gaussian broadening.

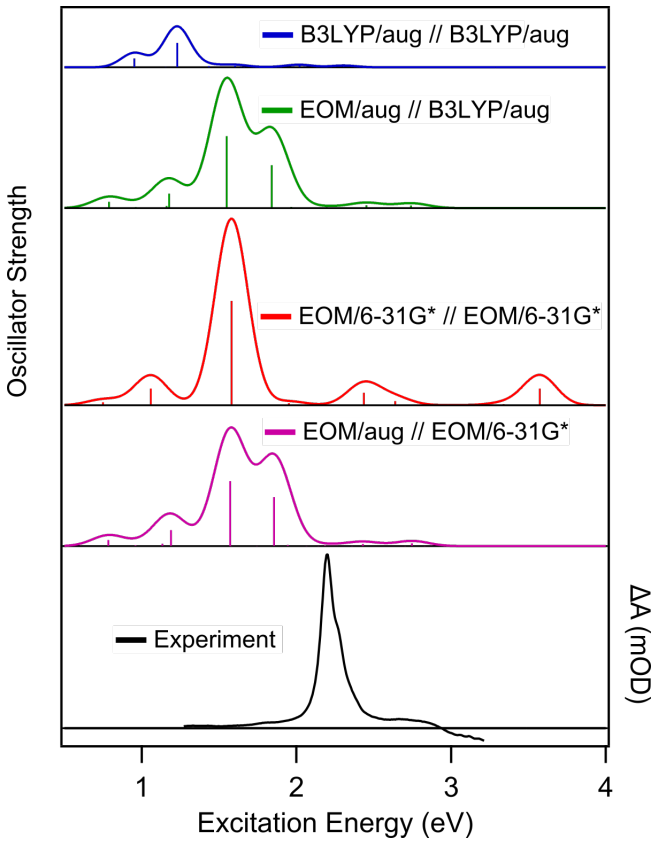


Figure S4: Comparison of calculated $S_n \leftarrow S_1$ transition energies and oscillator strengths for DPT with the experimental transient absorption spectrum. Calculated results were obtained using B3LYP/aug-cc-pVDZ, EOM-CCSD/6-31G*, or EOM-CCSD/aug-cc-pVDZ at the optimized geometry indicated for each spectrum. Calculated spectra include 0.25 eV Gaussian broadening. Values are from Tables S5-S10.

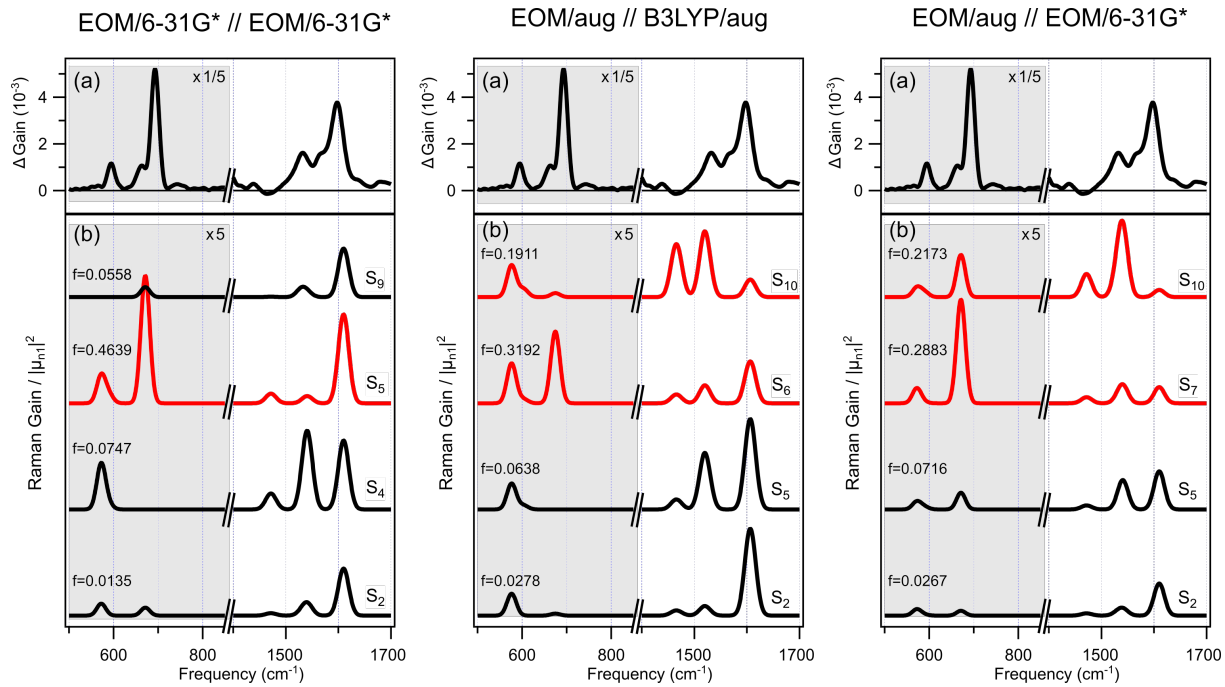


Figure S5: Comparison of simulated excited-state (S_1) resonance Raman spectra for different combinations of basis set and geometry. In each case, panel (a) shows experimental and panel (b) shows calculated resonance Raman spectra for DPT. Simulated results are from EOM-CCSD/6-31G* or EOM-CCSD/aug-cc-pVDZ using the optimized geometry and normal modes obtained from either EOM-CCSD/6-31G* or B3LYP/aug-cc-pVDZ, as indicated. Only states with $f \geq 0.013$ are shown. Simulated enhancements include 25 cm^{-1} Gaussian broadening, and include seven vibrations. Values are from Tables S7-S10.