Electronic Supplementary Material for:

Theoretical Study of Excited-State Dynamics of Hypercoordinated Carbon Molecule

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Table S1: Symmetry and harmonic frequency (ω) of the vibrational modes of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation at B3LYP/cc-pVDZ level of theory.

No.(Sym.)	Freq (in cm^{-1})	No.(Sym.)	Freq (in cm^{-1})
$\nu_1(a2)$	31.6031	$\nu_{61}(b2)$	1148.5701
$\nu_2(a2)$	44.5843	$\nu_{62}(b1)$	1155.3476
$\nu_3(b1)$	45.3309	$\nu_{63}(a2)$	1155.3691
$\nu_4(b1)$	74.0248	$\nu_{64}(a1)$	1155.5211
$\nu_5(b2)$	77.3328	$\nu_{65}(a1)$	1175.1323
$\nu_6(a2)$	83.6973	$\nu_{66}(b2)$	1179.0624
$\nu_7(b1)$	103.1632	$\nu_{67}(b1)$	1186.7340
$\nu_8(a2)$	113.7757	$\nu_{68}(a1)$	1190.6276
$\nu_9(a1)$	128.2130	$\nu_{69}(b2)$	1192.4994
$\nu_{10}(b2)$	129.7124	$\nu_{70}(a1)$	1211.5409
$\nu_{11}(b1)$	158.6749	$\nu_{71}(b2)$	1218.9835
$\nu_{12}(a1)$	180.6207	$\nu_{72}(b2)$	1276.6288
$\nu_{13}(b2)$	186.6579	$\nu_{73}(a1)$	1284.7263
$\nu_{14}(a2)$	187.9394	$\nu_{74}(a1)$	1303.7601
$\nu_{15}(b2)$	226.2238	$\nu_{75}(a1)$	1349.2596
$\nu_{16}(b1)$	231.2004	$\nu_{76}(b2)$	1357.1164
$\nu_{17}(a2)$	238.9861	$\nu_{77}(b2)$	1383.3315
$\nu_{18}(a2)$	250.8127	$\nu_{78}(b1)$	1395.1641
$\nu_{19}(b1)$	288.2786	$\nu_{79}(a1)$	1412.4506
$\nu_{20}(a1)$	307.5310	$\nu_{80}(b2)$	1415.1776
$\nu_{21}(b2)$	317.2665	$\nu_{81}(a1)$	1443.3354
$\nu_{22}(a1)$	321.7253	$\nu_{82}(a1)$	1448.6745
$\nu_{23}(a2)$	324.0607	$\nu_{83}(b2)$	1449.2728
$\nu_{24}(b1)$	328.3286	$\nu_{84}(b1)$	1450.4329
$\nu_{25}(a1)$	402.9798	$\nu_{85}(a1)$	1454.4699
$\nu_{26}(b1)$	462.2537	$\nu_{86}(a2)$	1460.2585
$\nu_{27}(a1)$	471.6552	$\nu_{87}(b1)$	1461.1050
$\nu_{28}(a2)$	501.9065	$\nu_{88}(b2)$	1463.0356
$\nu_{29}(b2)$	513.5067	$\nu_{89}(a2)$	1463.8208
$\nu_{30}(b2)$	518.0445	$\nu_{90}(b2)$	1467.5341
$\nu_{31}(b1)$	547.9998	$\nu_{91}(a1)$	1474.7227
$\nu_{32}(a1)$	552.3039	$\nu_{92}(b2)$	1476.5879
$\nu_{33}(b2)$	585.2234	$\nu_{93}(a1)$	1490.1824
$\nu_{34}(a2)$	613.3470	$\nu_{94}(a1)$	1504.7861
$\nu_{35}(b1)$	615.5136	$\nu_{95}(a1)$	1520.8748
$\nu_{36}(a1)$	615.8252	$\nu_{96}(b1)$	1560.8967
$\nu_{37}(b1)$	666.1830	$\nu_{97}(b2)$	1573.7883
$\nu_{38}(a1)$	674.5219	$\nu_{98}(a1)$	1608.6923
$\nu_{39}(b2)$	680.6648	$\nu_{99}(b2)$	1624.7587
$\nu_{40}(a2)$	755.8470	$\nu_{100}(b2)$	1666.6184
$\nu_{41}(b2)$	775.1942	$\nu_{101}(a1)$	1677.2264
$\nu_{42}(b1)$	777.3568	$\nu_{102}(b2)$	3028.0664
$\nu_{43}(a1)$	783.6739	$\nu_{103}(a1)$	3028.1806
$\nu_{44}(a2)$	819.0080	$\nu_{104}(b1)$	3077.5487
$\nu_{45}(b1)$	823.0350	$\nu_{105}(a1)$	3078.2887
$\nu_{46}(b1)$	892.9238	$\nu_{106}(a2)$	3108.6999
$\nu_{47}(a2)$	900.6654	$\nu_{107}(b1)$	3108.7588
$\nu_{48}(a1)$	925.4513	$\nu_{108}(b2)$	3161.2038
$\nu_{49}(b2)$	932.1095	$\nu_{109}(a1)$	3161.2348
$\nu_{50}(b1)$	940.8668	$\nu_{110}(a2)$	3183.6643
$\nu_{51}(b1)$	956.3633	$\nu_{111}(b2)$	3184.5817
$\nu_{52}(b2)$	983.2546	$\nu_{112}(a1)$	3185.2071
$\nu_{53}(a2)$	998.9667	$\nu_{113}(b2)$	3194.7933
$\nu_{54}(b1)$	1000.9021	$\nu_{114}(a1)$	3195.2200
$\nu_{55}(a1)$	1001.1289	$\nu_{115}(b1)$	3200.6384
$\nu_{56}(a1)$	1092.6004	$\nu_{116}(a1)$	3200.7559
$\nu_{57}(b2)$	1096.9290	$\nu_{117}(b2)$	3210.2171
$\nu_{58}(b2)$	1114.9977	$\nu_{118}(a1)$	3210.4486
$\nu_{59}(a1)$	1125.5836	$\nu_{119}(a1)$	3227.3118
$\nu_{60}(a2)$	1141.9471	$\nu_{120}(b2)$	3227.3324

Table S2: Ground-state (S_0) equilibrium geometry of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation optimized at B3LYP/cc-pVDZ level of theory.

	Coordinates (Angstrom)		
Atom	X	Y	Z
C	0.00000000	3.69339027	0.73100205
C	0.00000000	2.52368218	0.00507500
C	0.00000000	1.23813209	0.65701905
C	0.00000000	1.22775209	2.10079315
C	0.00000000	2.46323218	2.82303920
C	0.00000000	3.65741226	2.15422315
C	-0.00000000	-0.00000000	-0.04338400
C	-0.00000000	0.00000000	2.77211820
C	-0.00000000	-1.22775209	2.10079315
C	-0.00000000	-1.23813209	0.65701905
C	-0.00000000	-2.52368218	0.00507500
C	-0.00000000	-3.69339027	0.73100205
C	-0.00000000	-3.65741226	2.15422315
C	-0.00000000	-2.46323218	2.82303920
H	0.00000000	4.65768733	0.22498402
H	0.00000000	2.42983018	3.91362028
H	0.00000000	4.59995133	2.70390820
H	-0.00000000	0.00000000	3.86466528
H	-0.00000000	-4.65768733	0.22498402
H	-0.00000000	-4.59995133	2.70390820
H	-0.00000000	-2.42983018	3.91362028
O	-0.00000000	-2.45267918	-1.35743910
O	0.00000000	2.45267918	-1.35743910
C	0.00000000	-0.00000000	-1.53907111
O	-1.06578308	-0.00000000	-2.26320716
O	1.06578308	-0.00000000	-2.26320716
C	-2.38270817	0.00000000	-1.64403812
H	-3.07907322	0.00000000	-2.48883318
H	-2.50578518	-0.90557606	-1.03742607
H	-2.50578518	0.90557606	-1.03742607
C	2.38270817	-0.00000000	-1.64403812
H	2.50578518	0.90557606	-1.03742607
H	2.50578518	-0.90557606	-1.03742607
H	3.07907322	-0.00000000	-2.48883318
C	-0.00000000	-3.65959426	-2.12714615
H	-0.00000000	-3.34332924	-3.17729023
H	0.90144907	-4.25843131	-1.91874314
H	-0.90144907	-4.25843131	-1.91874314
C	0.00000000	3.65959426	-2.12714615
H	0.00000000	3.34332924	-3.17729023
H	-0.90144907	4.25843131	-1.91874314
H	0.90144907	4.25843131	-1.91874314

Table S3: Linear intrastate coupling parameters (κ) (in eV) for the singlet electronic states of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation obtained at TD-CAM-B3LYP/aug-cc-pVDZ level of theory. Excitation strength ($\kappa^2/2\omega^2$) obtained for each vibrational mode is shown in parenthesis.

a_1 mode (Freq, eV)	κ^{S_1}	κ^{S_2}
ν_9 (0.0159)	0.0089 (0.1578)	0.0526 (5.4779)
ν_{12} (0.0224)	-0.0027 (0.0074)	0.0036 (0.0130)
ν_{20} (0.0381)	0.0052 (0.0093)	-0.0094 (0.0306)
ν_{22} (0.0399)	0.0099 (0.0305)	0.0041 (0.0052)
ν_{25} (0.0500)	-0.0429 (0.3689)	-0.0186 (0.0688)
ν_{27} (0.0585)	0.0232 (0.0789)	-0.0203 (0.0600)
ν_{32} (0.0685)	-0.0005 (0.0000)	0.0376 (0.1507)
ν_{36} (0.0764)	-0.0419 (0.1505)	-0.0319 (0.0873)
ν_{38} (0.0836)	-0.0025 (0.0004)	0.0319 (0.0728)
ν_{43} (0.0972)	-0.0175 (0.0162)	0.0384 (0.0780)
ν_{48} (0.1147)	-0.0092 (0.0032)	-0.0063 (0.0015)
ν_{55} (0.1241)	-0.0033 (0.0003)	-0.0232 (0.0174)
ν_{56} (0.1355)	-0.0369 (0.0371)	-0.0250 (0.0170)
ν_{59} (0.1396)	-0.0196 (0.0099)	0.0896 (0.2061)
ν_{64} (0.1433)	0.0273 (0.0181)	0.0836 (0.1703)
ν_{65} (0.1457)	-0.0111 (0.0029)	0.0316 (0.0235)
ν_{68} (0.1476)	0.0088 (0.0018)	-0.0115 (0.0030)
ν_{70} (0.1502)	-0.0314 (0.0219)	-0.0172 (0.0065)
ν_{73} (0.1593)	0.0131 (0.0034)	0.0625 (0.0770)
ν_{74} (0.1616)	0.0863 (0.1425)	-0.0539 (0.0556)
ν_{75} (0.1673)	-0.0525 (0.0493)	-0.0709 (0.0898)
ν_{79} (0.1751)	-0.0778 (0.0986)	-0.2535 (1.0478)
ν_{81} (0.1790)	0.1580 (0.3896)	0.0553 (0.0478)
ν_{82} (0.1796)	-0.0473 (0.0346)	-0.0066 (0.0007)
ν_{85} (0.1803)	0.0326 (0.0163)	-0.0012 (0.0000)
ν_{91} (0.1828)	0.0096 (0.0014)	0.0277 (0.0115)
ν_{93} (0.1848)	-0.0444 (0.0288)	0.0278 (0.0113)
ν_{94} (0.1866)	-0.0333 (0.0160)	0.0229 (0.0075)
ν_{95} (0.1886)	-0.0348 (0.0170)	-0.2218 (0.6914)
ν_{98} (0.1995)	-0.1887 (0.4473)	-0.1676 (0.3530)
ν_{101} (0.2080)	0.0044 (0.0002)	-0.0027 (0.0001)
ν_{103} (0.3754)	0.0041 (0.0001)	0.0001 (0.0000)
ν_{105} (0.3817)	0.0001 (0.0000)	0.0314 (0.0034)
ν_{109} (0.3919)	0.0000 (0.0000)	-0.0047 (0.0001)
ν_{112} (0.3949)	0.0102 (0.0003)	0.0003 (0.0000)
ν_{114} (0.3962)	0.0007 (0.0000)	-0.0009 (0.0000)
ν_{116} (0.3968)	0.0020 (0.0000)	-0.0112 (0.0004)
ν_{118} (0.3980)	0.0014 (0.0000)	-0.0005 (0.0000)
ν_{119} (0.4001)	0.0095 (0.0003)	0.0047 (0.0001)

Table S4: Linear interstate coupling parameters (λ) (in eV) computed along a_2 modes for singlet electronic states of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation obtained at TD-CAM-B3LYP/aug-cc-pVDZ level of theory. Excitation strength ($\lambda^2/2\omega^2$) obtained for each vibrational mode is shown in paranthesis.

a_2 mode (Freq, eV)	$\lambda_{S_1-S_2}$
ν_1 (0.0039)	0.0812 (216.4904)
ν_2 (0.0055)	0.0143 (3.3728)
ν_6 (0.0104)	0.0042 (0.0813)
ν_8 (0.0141)	0.0323 (2.6190)
ν_{14} (0.0233)	0.0187 (0.3215)
ν_{17} (0.0296)	0.0721 (2.9678)
ν_{18} (0.0311)	0.0019 (0.0019)
ν_{23} (0.0402)	0.0188 (0.1099)
ν_{28} (0.0622)	0.0201 (0.0520)
ν_{34} (0.0760)	0.0277 (0.0663)
ν_{40} (0.0937)	0.0382 (0.0830)
ν_{44} (0.1015)	0.0281 (0.0384)
ν_{47} (0.1117)	0.0312 (0.0389)
ν_{53} (0.1239)	0.0264 (0.0227)
ν_{60} (0.1416)	—
ν_{63} (0.1432)	—
ν_{86} (0.1810)	0.0098 (0.0015)
ν_{89} (0.1815)	—
ν_{106} (0.3854)	—
ν_{110} (0.3947)	—

Table S5: MCTDH details of normal modes combination, size of the primitive and single particle bases used in the S_1-S_2 vibronic dynamics of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation.

Normal modes	Primitive basis	SPF
(ν_1, ν_9, ν_{25}, ν_{40}, ν_{98})	(9, 8, 7, 6, 7)	[8, 10]
(ν_2, ν_{27}, ν_{32}, ν_{34}, ν_{95})	(8, 5, 7, 6, 8)	[6, 8]
(ν_6, ν_{28}, ν_{36}, ν_{38}, ν_{81})	(6, 6, 6, 6, 8)	[7, 6]
(ν_8, ν_{23} ν_{43}, ν_{59}, ν_{79})	(8, 7, 6, 7, 8)	[7, 9]
(ν_{14}, ν_{17}, ν_{64}, ν_{73}, ν_{74}, ν_{75})	(7, 8, 7, 6, 7, 6)	[7, 8]

Table S6: S_1-S_2 minimum energy conical intersection (MECI) geometry of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation optimized at TD-CAM-B3LYP/aug-cc-pVDZ level of theory using GRRM.

	Coordinates (Angstrom)		
Atom	X	Y	Z
C	-0.000208792713	3.703740268953	0.750168733598
C	-0.000791692297	2.521341077847	-0.007769772460
C	-0.000438206966	1.241229574776	0.626400275406
C	0.000136290826	1.228665736782	2.060694355192
C	0.000290024284	2.432450209142	2.793487956454
C	0.000159249543	3.655623416338	2.139122714878
C	-0.000312360151	-0.000003463007	-0.090049973859
C	0.000641040593	0.000000617721	2.738544605728
C	0.000462702099	-1.228666707126	2.060700000695
C	-0.000077399788	-1.241234560410	0.626406882268
C	-0.000270766667	-2.521342506374	-0.007761985233
C	0.000120103839	-3.703744069358	0.750173917479
C	0.000706391710	-3.655625592810	2.139127537892
C	0.000967227357	-2.432452219239	2.793493207024
H	-0.000262037980	4.667740204938	0.251565434162
H	0.000650758692	2.389927044361	3.881083619668
H	0.000391567378	4.582372605727	2.709759740274
H	0.001140941425	0.000003278081	3.827833390590
H	-0.000015417080	-4.667744088447	0.251570965899
H	0.001056590439	-4.582374187189	2.709766063291
H	0.001531631688	-2.389929248071	3.881088926702
O	-0.000862571198	-2.533086118600	-1.344714113423
O	-0.001369891675	2.533090008474	-1.344719653442
C	0.000553181006	-0.000006181987	-1.559202836037
O	-1.086295623356	-0.000270102274	-2.314411375033
O	1.088719413669	0.000318014404	-2.312518838531
C	-2.366446708248	0.000583016716	-1.681032097778
H	-3.092309236230	0.000169369124	-2.495736609784
H	-2.496268256373	-0.898077357678	-1.067265031085
H	-2.495723690918	0.900280793051	-1.068675808537
C	2.368109878340	-0.000585243880	-1.677688993372
H	2.497307139368	0.898033459701	-1.063712468598
H	2.496741141209	-0.900308630799	-1.065218405581
H	3.094829038486	-0.000137387723	-2.491658487605
C	-0.001469041311	-3.772031154709	-2.059072879165
H	-0.002060397710	-3.493338435215	-3.114318412237
H	0.899724960804	-4.353004230750	-1.828307441182
H	-0.902520321184	-4.352799575049	-1.827183642656
C	-0.001597833036	3.772035894728	-2.059076763161
H	-0.001630230057	3.493343502762	-3.114322285585
H	-0.902777196559	4.352802822404	-1.827678447425
H	0.899468398743	4.353010144636	-1.827820155406

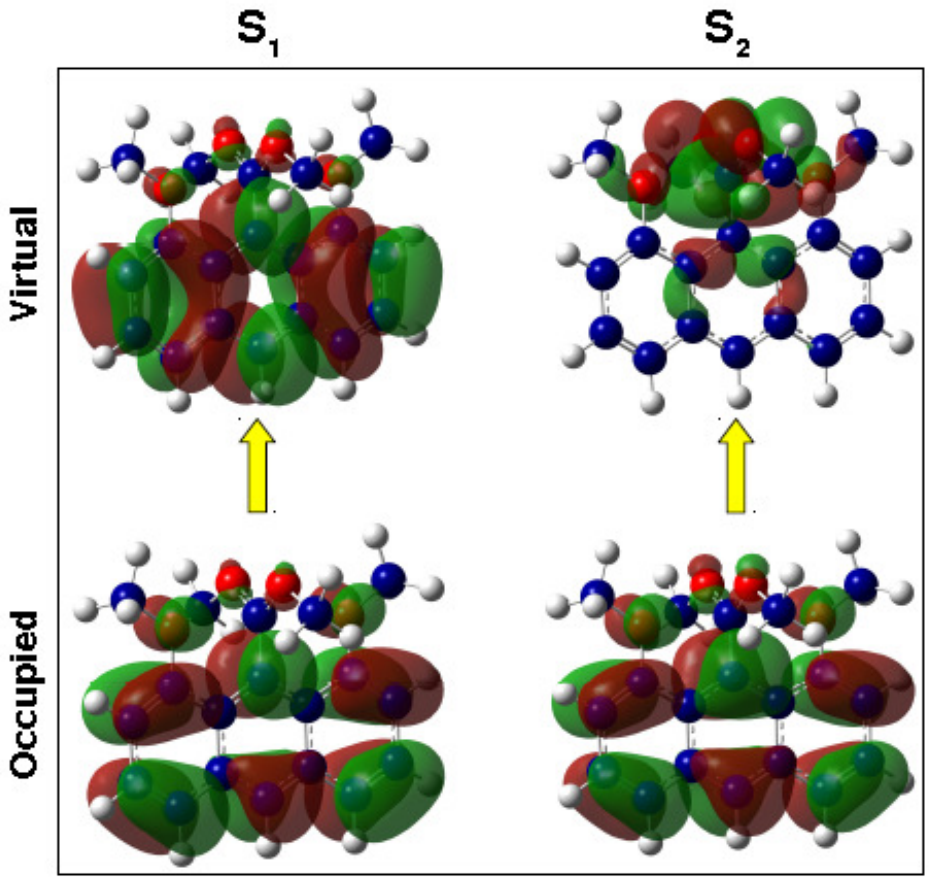

Figure S1: Natural transition orbitals (NTOs) showing major orbitals contributing towards excitation of 1,8-dimethoxy-9-dimethoxymethylanthracene monocation obtained at TD-CAM-B3LYP/aug-cc-pVDZ level of theory.