Supporting Information For:

Efficient Treatment of Large Active Spaces through Multi-GPU Parallel Implementation of Direct Configuration Interaction

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Benchmark Timing Supporting Information

To allow the reader to make meaningful comparisons and draw his/her own conclusions, we have provided detailed timing information for σ and 2-RDM formation for each of the hardware configurations presented in the main text. Where applicable, we have also given speedups relative to the single GPU calculation.

Table S1. Timings for σ formation for CAS-(18,18)-CI/6-31G** on ethylene dimer. Results using between one and six 1080Ti GPUs are shown. Minimum timings are shown, with average timings over all formations in parentheses.

Step	Time (s)					
	1 GPU	**2 GPUs**	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**
C/σ Trans.	52.49 (68.13)	32.31 (36.67)	30.54 (25.42)	21.73 (25.95)	20.28 (24.35)	18.65 (25.04)
GPU Mem. Ops.	524.80 (526.61)	279.57 (308.63)	199.88 (215.44)	183.51 (201.72)	154.26 (165.69)	143.82 (167.97)
$C_\alpha / C_\beta / \sigma_\alpha / \sigma_\beta$	574.17 (601.42)	307.33 (343.55)	196.59 (224.56)	151.69 (188.95)	129.47 (148.54)	123.78 (175.00)
$C_{\alpha\beta} / \sigma_{\alpha\beta}$	7.77 (8.14)	4.49 (4.96)	2.84 (3.18)	2.25 (2.74)	1.88 (2.16)	1.78 (2.37)
σ_{1e} **(DGEMV)**	16.28 (16.29)	8.13 (8.14)	5.42 (5.42)	4.06 (4.06)	3.25 (3.25)	2.71 (2.71)
D Kernels	59.98 (59.99)	29.61 (29.97)	19.95 (19.97)	14.49 (14.98)	11.97 (11.99)	9.87 (10.00)
$\mathrm{E} = (ij \mid kl) \times \mathrm{D}$ **(DGEMM)**	1572.85 (1573.88)	790.59 (792.09)	516.54 (525.10)	385.14 (394.19)	309.19 (314.91)	256.55 (262.43)
σ Kernels	84.00 (84.03)	41.37 (41.87)	27.89 (27.91)	20.24 (20.94)	16.71 (16.74)	13.80 (13.97)
σ Scatter	1.99 (2.81)	4.62 (12.58)	5.68 (12.54)	6.68 (7.51)	7.89 (15.67)	10.88 (14.94)
Tot. σ Time	3010.67 (3045.03)	1626.52 (1681.51)	1122.88 (1153.85)	899.67 (939.11)	761.66 (779.14)	720.99 (775.38)

Table S2. Speedups for σ formation for CAS-(18,18)-CI/6-31G** on ethylene dimer. Results using between one and six 1080Ti GPUs are shown. Average speedups are shown, with maximum speedups over all formations in parentheses.

Step	**Time (s)**				
	2 GPUs	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**
C/σ Trans.	1.86x (1.62x)	2.23x (2.06x)	2.62x (2.41x)	2.80x (2.59x)	2.72x (2.81x)
GPU Mem. Ops.	1.71x (1.88x)	2.44x (2.63x)	2.61x (2.86x)	3.18x (3.40x)	3.14x (3.65x)
$C_\alpha / C_\beta / \sigma_\alpha / \sigma_\beta$	1.75x (1.87x)	2.68x (2.92x)	3.18x (3.79x)	4.05x (4.43x)	3.44x (4.64x)
$C_{\alpha\beta} / \sigma_{\alpha\beta}$	1.64x (1.73x)	2.56x (2.74x)	2.97x (3.45x)	3.77x (4.13x)	3.43x (4.37x)
σ_{1e} **(DGEMV)**	2.00x (2.00x)	3.00x (3.01x)	4.01x (4.01x)	5.01x (5.01x)	6.01x (6.01x)
D Kernels	2.00x (2.03x)	3.00x (3.01x)	4.00x (4.13x)	5.00x (5.01x)	6.00x (6.08x)
$\mathrm{E} = (\mathrm{ij}\|\mathrm{kl}) \times \mathrm{D}$ **(DGEMM)**	1.99x (1.99x)	3.00x (3.05x)	3.99x (4.09x)	5.00x (5.09x)	6.00x (6.13x)
σ Kernels	2.01x (2.03x)	3.01x (3.01x)	4.01x (4.15x)	5.02x (5.03x)	6.01x (6.09x)
Tot. σ Time	1.81x (1.85x)	2.64x (2.68x)	3.24x (3.35x)	3.91x (3.95x)	(3.93x) (4.17x)

Table S3. Timings for 2-RDM formation for CAS-(18,18)-CI/6-31G** on ethylene dimer. Results using between one and six 1080Ti GPUs are shown. Minimum timings are shown, with average timings over all formations in parentheses. 2-RDM average timings.

Step	Time (s)					
	1 GPU	**2 GPUs**	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**
Mem.	1294.48 (1294.48)	878.38 (878.76)	562.07 (566.03)	470.91 (477.50)	465.73 (470.44)	331.38 (364.11)
GPU Kernels	119.62 (119.62)	59.04 (59.75)	39.81 (39.83)	28.91 (29.87)	23.88 (23.91)	19.68 (19.92)
DGEMM	1857.52 (1857.53)	928.66 (928.93)	606.07 (614.90)	454.22 (461.73)	363.13 (369.27)	303.34 (308.50)
DGEMV	23.46 (23.46)	15.80 (15.81)	10.40 (10.49)	7.80 (7.89)	6.25 (6.31)	5.22 (5.27)
Tot. 2-RDM	3390.58 (3390.59)	1941.84 (1941.84)	1285.81 (1285.81)	1029.17 (1029.17)	910.36 (910.36)	738.37 (738.37)

Table S4. Speedups for 2-RDM formation for CAS-(18,18)-CI/6-31G** on ethylene dimer. Results using between one and six 1080Ti GPUs are shown. Average speedups are shown, with maximum speedups over all formations in parentheses.

Step	Time (s)				
	2 GPUs	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**
Mem.	1.47x (1.47x)	2.29x (2.30x)	2.71x (2.75x)	2.75x (2.78x)	3.55x (3.91x)
GPU Kernels	2.00x (2.03x)	3.00x (3.00x)	4.00x (4.14x)	5.00x (5.01x)	6.00x (6.08x)
DGEMM	2.00x (2.00x)	3.02x (3.06x)	4.02x (4.09x)	5.03x (5.12x)	6.02x (6.12x)
DGEMV	1.48x (1.48x)	2.24x (2.26x)	2.97x (3.01x)	3.72x (3.75x)	4.45x (4.49x)
Tot. 2-RDM	1.75x (1.75x)	2.64x (2.64x)	3.29x (3.29x)	3.72x (3.72x)	4.59x (4.59x)

Table S5. Timings for σ formation for CAS-(16,17)-CI/6-31G** on ethylene. Results using between one and eight P100 GPUs are shown. Minimum timings are shown, with speedups over single GPU calculation in parentheses.

Step	**Time (s)**							
	1 GPU	**2 GPUs**	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**	**7 GPUs**	**8 GPUs**
GPU Mem. Ops.	---	2.47	1.65	2.05	1.90	1.77	1.18	1.76
σ_{1e} **(DGEMV)**	2.43	5.32	3.54	2.66	2.12	1.77	1.51	1.32
D Kernels	5.68	2.81 (2.02x)	1.85 (3.07x)	1.37 (4.15x)	1.14 (4.98x)	0.90 (6.31x)	0.81 (7.01x)	0.66 (8.61x)
$E = (ij\|kl) \times D$ **(DGEMM)**	26.52	12.91 (2.05x)	8.61 (3.09x)	6.45 (4.11x)	5.15 (5.15x)	4.30 (6.17x)	3.69 (7.19x)	3.23 (8.21x)
σ **Kernels**	6.75	3.34 (2.02x)	2.20 (3.07x)	1.64 (4.12x)	1.35 (5.00x)	1.07 (6.30x)	0.96 (7.03x)	0.78 (8.65x)
σ **Scatter**	---	0.93	1.25	1.20	1.44	1.54	1.74	1.91
Total σ **Time**	48.89	25.76 (1.90x)	18.81 (2.60x)	15.20 (3.22x)	13.98 (3.50x)	12.00 (4.07x)	11.29 (4.33x)	10.89 (4.49x)

Table S6. Timings for 2-RDM formation for CAS-(16,17)-CI/6-31G** on ethylene. Results using between one and eight P100 GPUs are shown.

Step	Time (s)							
	1 GPU	**2 GPUs**	**3 GPUs**	**4 GPUs**	**5 GPUs**	**6 GPUs**	**7 GPUs**	**8 GPUs**
Mem.	8.25	4.98	3.70	3.46	3.64	3.49	3.06	3.87
GPU Kernels	11.40	5.63	3.72	2.75	2.28	1.79	1.62	1.31
DGEMM	50.82	25.41	16.94	12.71	10.15	8.47	7.26	6.35
DGEMV	10.63	5.32	3.55	2.66	2.12	1.77	1.51	1.33
Tot. 2-RDM	81.25	41.63	28.55	21.86	18.36	15.77	13.95	13.61

Diphenyl Acetylene Absorption Spectra Supporting Information

We present here molecular and natural orbitals computed using FOMO-CAS-(16,16)-CI and SA-5-CAS-(16,16)-SCF, respectively, computed at the Franck-Condon point.

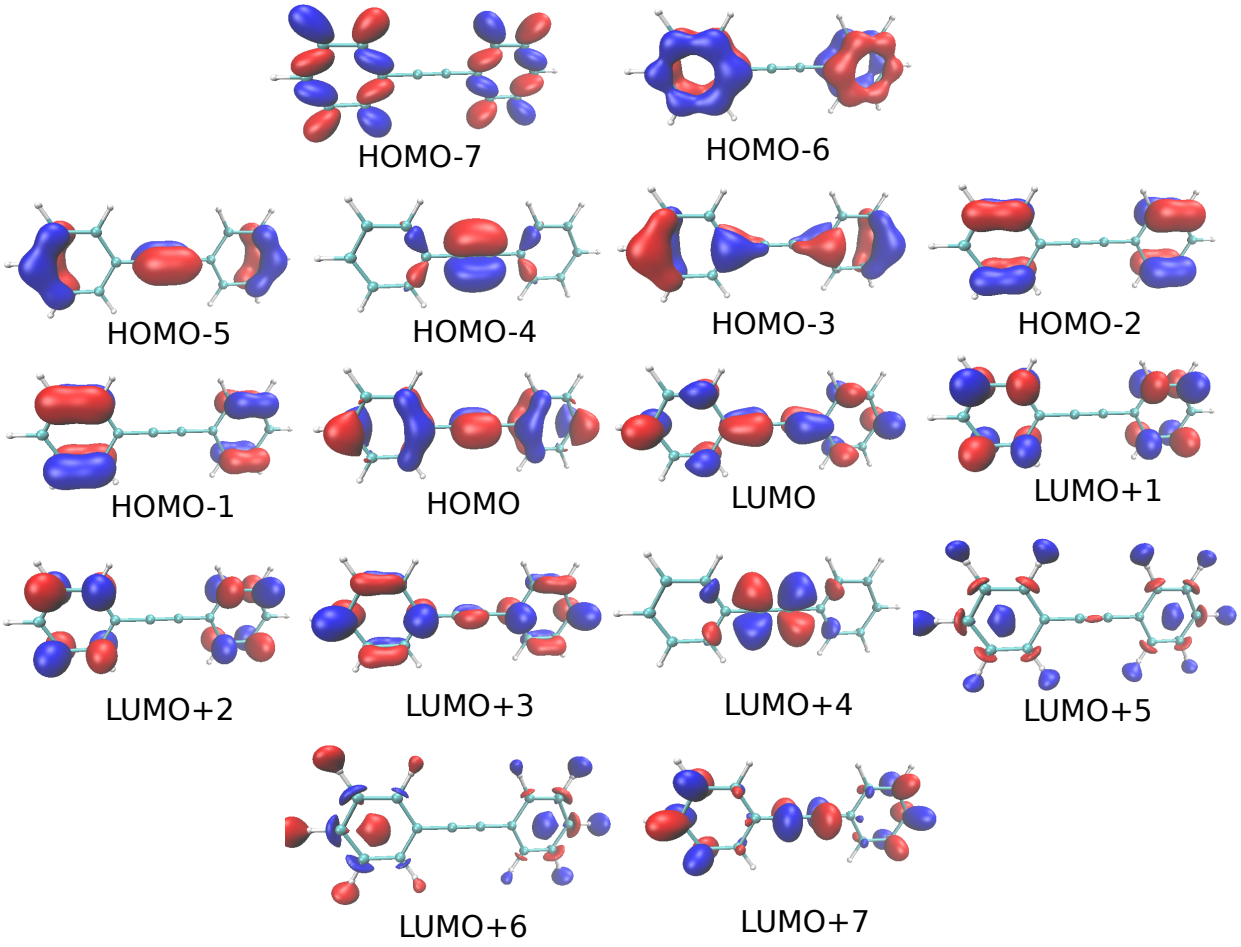

Figure S1. FOMO-CAS-(16,16)-CI/6-31G* diphenyl acetylene molecular orbitals.

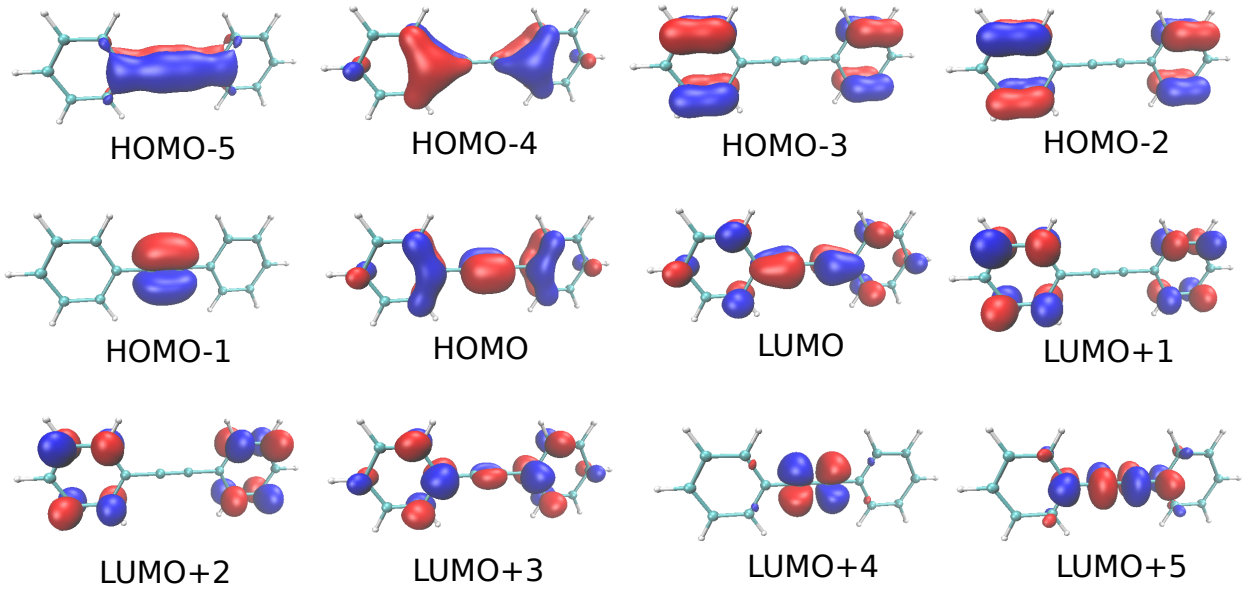

Figure S2. SA-5-CAS-(12,12)-SCF/6-31G* diphenyl acetylene natural orbitals.

Cartesian Coordinates for Molecular Geometries

Diphenyl acetylene optimized at PBE0/6-31G.*

24

C	-0.4790840216	0.0004750194	0.0322667735
C	-0.4652993198	1.2159870079	0.0333333992
C	-0.4874473833	-1.4221971817	0.0256449307
C	-0.4597786926	2.6386889427	0.0372253377
C	-1.6975835214	-2.1311216091	-0.0595057199
C	0.7175709250	-2.1405529373	0.1015973453
C	-1.6970627995	-3.5197668241	-0.0717801592
C	0.7086491885	-3.5289340629	0.0890241795
C	-0.4963265921	-4.2233543763	0.0013696684
C	0.7498040090	3.3505798875	0.1018793046
C	-1.6677890252	3.3538356173	-0.0203839368
C	0.7459698213	4.7392657432	0.1107561892
C	-1.6621387594	4.7423003233	-0.0110746075
C	-0.4574457478	5.4399108468	0.0549469877
H	-2.6311819098	-1.5796139262	-0.1150592079
H	-2.6395240198	-4.0566708328	-0.1380289291
H	1.6477374726	-4.0724538402	0.1483933095
H	1.6537536573	-1.5950343045	0.1707539993
H	-2.6036000414	2.8053123337	-0.0708065976
H	-2.6033813519	5.2835159984	-0.0546752094
H	1.6854679104	2.8012434152	0.1455616592
H	1.6879949389	5.2783055318	0.1622217391
H	-0.4562459824	6.5263929802	0.0641082311
H	-0.4996587556	-5.3097137557	-0.0087686864

Ethylene dimer used for benchmark calculations.

12

C	0.35673483	-0.05087227	-0.47786734
C	1.61445821	-0.06684947	-0.02916681
H	-0.14997206	0.87780529	-0.62680155
H	-0.16786485	-0.95561368	-0.69426370
H	2.15270896	0.84221076	0.19314809
H	2.16553127	-0.97886933	0.15232587
C	6.35673483	-0.05087227	-0.47786734
C	7.61445821	-0.06684947	-0.02916681
H	5.85000000	0.87780529	-0.62680155
H	5.84000000	-0.95561368	-0.69426370
H	8.15270896	0.84221076	0.19314809
H	8.16553127	-0.97886933	0.15232587