Supporting information for:

Evaluating the GW approximation with CCSD(T) for charged excitations across the oligoacenes

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This document contains:

1. CCSD(T) references.

    - FPA: We detail the focal point analysis (FPA)[S1] used in this work.

    - Our BTE: We analyze the FPA done in Ref. S1 and explain how we obtain the best theoretical estimates (BTE).

2. Excitation energies. We list the ionization potential and electron affinity energies calculated within DFT and *GW* in this work.

3. OTRSH convergence. We show convergence studies of charged excitations within OTRSH.

4. Geometries. We provide the relaxed geometries of the oligoacenes used in this work.

1 CCSD(T) references

Here we describe how we have calculated the best theoretical estimates within CCSD(T).

1.1 Focal point analysis

We begin by detailing the focal point analysis (FPA) of Ref. S1, which is also used in this work.

The CCSD(T) energies are obtained from HF total energies and adding corrections to the correlation at the second-, third- and partial fourth-order Møller-Plesset levels (MP2, MP3 and MP4SDQ). Further improvements are added from coupled-cluster calculations including singlets and doubles (CCSD) and a perturbative estimate of the triples [CCSD(T)].

In the FPA, the complete basis set (CBS) limit is extrapolated using Dunning basis sets of increasing size. For the HF energy at the CBS limit, E_∞^{HF}, is extrapolated using the Feller's

formula:

$$E^{\mathrm{HF}}(l) = E^{\mathrm{HF}}_{\infty} + Ae^{-Bl}, \tag{1}$$

where l equals 2, 3, or 4, corresponding to the cc-pVDZ, cc-pVTZ and cc-pVQZ basis set respectively, and A and B are constants. Later the post-HF energy corrections, E^{corr}, at the MP2, MP3, MP4, CCSD and CCSD(T) levels, are extrapolated using the Schwartz formula:

$$E^{\mathrm{corr.}}(l) = E^{\mathrm{corr.}}_{\infty} + \frac{C}{(l + \frac{1}{2})^4} + \frac{D}{(l + \frac{1}{2})^6}, \tag{2}$$

where C and D are constants. In the following, all post-HF energies are reported as corrections rather than as absolute energies. So, for example, to obtain the IP energy at the MP3 level and DZ basis for anthracene using the data in the following tables, one needs to add HF + MP2 + MP3 in the DZ column; to obtain the IP energy at the MP3 level and TZ basis, one needs to further add the HF energy difference from the TZ and DZ basis (TZ−DZ) and the corresponding differences for the MP2 and MP3 corrections, i.e. $\mathrm{IP}^{\mathrm{MP3}}(\mathrm{TZ})$ = HF(DZ) + HF(TZ-DZ) + MP2(DZ) + MP2(TZ-DZ) + MP3(DZ) + MP3(TZ-DZ).

1.2 Our best theoretical estimates

We decided to update the reference values of Ref. S1 because when making comparisons to them, we noticed a significant break in the trend at hexacene. For any given method, e.g. G_0W_0@PBE0 or evGW@BHLYP we observed a more or less constant difference between the method being tested and the CCSD(T) value from Ref. S1 except for hexacene. We observed that if the reference CCSD(T) value was ∼0.1 eV too high, this would explain the break in the trend.

Upon examining the focal point analysis (FPA) undertaken in Ref. S1, we found one assumption in particular, that by itself could account for 0.1 eV extra in the CCSD(T) value for hexacene. The culprit assumption was that the MP3 contribution to the CCSD(T) energy was converged well enough at the DZ level, but considering the differences between

TZ and DZ MP3 corrections for acenes, see Table S1, it seems that this assumption leads to an error of ~0.1 eV for hexacene. For instance, based on the trend in TZ − DZ column of Table S1, we deduce that the TZ correction over the DZ energy for hexacene is -0.1 eV rather than 0.0 eV, as used in Ref. S1.

Table S1: We show the MP3 corrections over MP2 energies, ΔE^{MP3}, calculated with the DZ basis, as well as the differences between the corrections at the TZ and DZ levels (TZ − DZ) and between the corrections at the QZ and TZ levels (QZ − TZ). All energies are taken from Ref. S1. Note: italicized numbers were assumed, not calculated.

	ΔE^{MP3} (eV)		
	DZ	TZ − DZ	QZ − TZ
Benzene	-0.29	-0.05	-0.02
Naphthalene	-0.24	-0.06	-0.02
Anthracene	-0.23	-0.07	*0.0*
Tetracene	-0.26	-0.08	*0.0*
Pentacene	-0.30	-0.09	*0.0*
Hexacene	-0.36	*0.0*	*0.0*

Furthermore, based upon the trend in the QZ −TZ corrections for benzene and naphthalene (see Table S1), we may as well assume that the correction at the QZ level for anthracene through hexacene is -0.02 eV rather than 0.0 eV as used in Ref. S1. Making similar tables at the HF, MP2, MP4, CCSD, and CCSD(T) levels with the data reported in Ref. S1, we find that all assumptions and extrapolations look quite reasonable, except another very small one: based on the MP4 correction trend on shorter acenes (see Table S2), the TZ − DZ MP4 correction over the MP3 energy for hexacene is better estimated as -0.02 eV rather than the 0.0 eV taken by Ref. S1.

Using the energies calculated in Ref. S1 and changing the MP3 and MP4 energy corrections as described above gives an updated list of CCSD(T) reference values, which we refer to as "corrected Deleuze values". In order to further check these numbers, we repeated the work of Ref. S1, closely following the methodology described in their paper, and detailed in Section 1.1. The results are summarized next; the BTE is taken to be an average of the corrected Deleuze values with our independently determined values. The uncertainty is given

Table S2: We show MP4 corrections over MP3 energies, ΔE^{MP4}, calculated with the DZ basis set and the differences between the corrections at increasing basis set size, as in Table S1. All energies are taken from Ref. S1. Note: italicized numbers were assumed, not calculated.

	ΔE^{MP4} (eV)		
	DZ	TZ $-$ DZ	QZ $-$ TZ
Benzene	-0.04	-0.02	0.00
Naphthalene	-0.05	-0.02	-0.01
Anthracene	-0.04	-0.02	*0.0*
Tetracene	-0.06	-0.02	*0.0*
Pentacene	-0.09	-0.02	*0.0*
Hexacene	-0.12	*0.0*	*0.0*

based upon the disagreement in these sets of numbers; this is not an absolute uncertainty, but an uncertainty within the computational method: CCSD(T) extrapolating to the CBS limit.

Table S3: CCSD(T) energies in the CBS limit for the acenes (in eV)

	Ref. S1	Updated Ref. S1	Present Work	Best Theoretical Estimate
Benzene	9.45	9.45	9.44	9.44 $\pm$ 0.01
Naphthalene	8.24	8.24	8.26	8.25 $\pm$ 0.01
Anthracene	7.47	7.45	7.51	7.48 $\pm$ 0.03
Tetracene	6.95	6.93	6.99	6.96 $\pm$ 0.03
Pentacene	6.57	6.55	6.61	6.58 $\pm$ 0.03
Hexacene	6.43	6.31	6.32	6.32 $\pm$ 0.03

We now give the details of our calculations. All work was carried out within the Gaussian G09 E.01 software package.[S2] A notable difference between the present work and that of Ref. S1 is that their calculations utilize unrestricted HF, while ours are based upon restricted HF. We made this choice to avoid spin contamination; however, we observe that even though the unrestricted wavefunctions possess spin contamination, the post Hartree Fock (HF) corrections largely eliminate energy differences, so that by the time an MP4 correction is incrementally added, the difference in the IP or EA energies using the two different HF starting points is less than 0.05 eV. To illustrate, consider the energy differences between HF and post-HF corrections between Ref. S1 (unrestricted) and the present work (restricted)

for naphthalene:

Table S4: $\Delta E^{\mathrm{UHF-RHF}}$: difference between the resulting energies calculated relying on restricted HF (Present Work) minus those relying on unrestricted HF (taken from Ref. S1), for naphthalene. All energies are in units of eV.

| | $\Delta E^{\mathrm{UHF-RHF}}$ (eV) | | | | |
	cc-pVDZ	aug-cc-pVDZ	cc-pVTZ	cc-pVQZ	cc-pV∞Z
HF	0.34	0.35	0.36	0.36	0.36
+MP2	-0.71	-0.75	-0.76	-0.78	-0.75
+MP3	0.30	0.32	0.33	0.33	0.33
+MP4(SDQ)	0.08	0.09	0.10	0.11	0.10
+CCSD	0.01	0.02	0.01	0.00	0.01
+CCSD(T)	-0.03	-0.04	-0.04	-0.03	-0.03
Total	-0.01	-0.01	-0.01	0.00	0.02

In Table S5, we report the energies we have calculated in such a way as to make assumptions, extrapolations, and trends clear. A white background indicates that no extrapolation or assumptions were made outside of those already inherent to the theory. A purple cell indicates that the presented value involves either Feller or Schwartz extrapolation as described above, while a yellow cell indicates that the presented value was linearly extrapolated based on the trend in that column. We assert that because trends are clearly visible. In contrast, a different choice is made in Ref. S1 that whenever it was unfeasible to calculate, e.g. MP3 at the QZ level, MP3 at the TZ level was taken at face value as converged well-enough. As has already been discussed, this is reasonable in most cases, but not in the case of hexacene.

Table S5: IP energies of the acenes calculated at the UHF level and correlation energy corrections. We show the results using the DZ basis set and energy differences from calculations with different basis sets, following the convention in Table S1. Colored cells indicate an extrapolation, as explained in the text.

UHF Ionization Potential Energies (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	8.232	0.003	0.008	0.007
Naphthalene	7.250	0.015	0.017	0.011
Anthracene	6.458	0.016	0.019	0.028
Tetracene	5.910	0.017	0.015	0.013
Pentacene	5.510	0.016	0.012	0.006
Hexacene	5.153	0.015	0.017	0.015

+MP2 Ionization Energy corrections (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	0.960	0.199	0.062	0.038
Naphthalene	0.627	0.194	0.064	0.043
Anthracene	0.615	0.203	0.074	0.046
Tetracene	0.597	0.207	0.083	0.065
Pentacene	0.602	0.222	0.083	0.057
Hexacene	0.653	0.221	0.083	0.069

+MP3 Ionization Energies (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	-0.101	-0.038	-0.009	-0.001
Naphthalene	0.056	-0.028	-0.014	-0.011
Anthracene	0.082	-0.031	-0.014	-0.003
Tetracene	0.112	-0.031	-0.014	-0.017
Pentacene	0.129	-0.031	-0.014	-0.009
Hexacene	0.122	-0.031	-0.014	-0.023

+MP4 Ionization Energies (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	-0.025	-0.012	-0.008	-0.006
Naphthalene	0.034	-0.006	-0.003	-0.005
Anthracene	0.042	-0.005	-0.003	-0.013
Tetracene	0.053	-0.004	-0.003	0.005
Pentacene	0.056	-0.003	-0.003	-0.009
Hexacene	0.052	-0.002	-0.003	0.003

+CCSD Ionization Energies (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	0.027	-0.004	0.003	0.007

Naphthalene	0.005	-0.012	-0.002	0.003
Anthracene	-0.004	-0.014	-0.002	0.001
Tetracene	-0.011	-0.016	-0.002	0.000
Pentacene	-0.017	-0.016	-0.002	0.015
Hexacene	-0.019	-0.016	-0.002	-0.013

+CCSD(T) Ionization Energies (eV)

	DZ	TZ - DZ	QZ - TZ	∞Z - QZ
Benzene	0.039	0.043	0.012	0.003
Naphthalene	-0.022	0.039	0.000	0.002
Anthracene	-0.021	0.039	0.000	-0.002
Tetracene	-0.017	0.039	0.000	-0.006
Pentacene	-0.017	0.039	0.000	-0.015
Hexacene	-0.017	0.039	0.000	0.017

2 Excitation energies

In this section, we list the excitation energies calculated within DFT and GW in this work and we briefly comment on the performance of a non-tuned RSH starting point for GW.

2.1 Energetics from a non-tuned RSH starting point for GW

Recently, in Ref. S3 G_0W_0@RSH with fixed α and γ parameters have been benchmarked against CCSD(T) references and yielded highly accurate energetics on representative organic molecules. Inspired by these findings, we have also considered the RSH starting point for G_0W_0 with $\alpha = 0.2$ (as in OTRSH) and two arbitrary values of α; we set $\alpha = 0.4$ Bohr^{-1} (the default in LC-ωPBE) and 0.25 Bohr^{-1} for the sake of comparison. As shown in Tables S7 and S10, in agreement with Ref. S3, G_0W_0@RSH with fixed parameters can perform as precisely as G_0W_0@OTRSH for this set of excitations; in fact, G_0W_0@RSH with $\alpha = 0.2$ and $\gamma = 0.25$ Bohr^{-1} predicts IP and EA energetics with relatively low MADs of 0.2 and 0.3 eV, respectively.

2.2 List of excitation energies

In the following tables, we list the excitation energies calculated with DFT and GW in this work. The lowest unoccupied molecular orbitals (LUMO) of benzene and naphthalene are unbound for most DFT and GW schemes and difficult to tag due to the fact that they are energetically above a large (infinite, in principle) number of delocalized near-Fermi vacuum levels. Hence, in this work, unbound LUMOs are identified by projecting the calculated MOs onto the LUMOs obtained with DFT-PBE (which are bound) and then taking the MOs with highest LUMO overlap ($> 90\%$). Alternatively, the LUMOs can be tagged by a direct comparison to the LUMO densities shown in Ref. S4 or by comparing to the LUMO densities obtained with the non-augmented Dunning basis sets.

Table S6: Ionization potential energies calculated with DFT (in eV).

| | DFT | | | | CCSD(T) |
| | RSH ($\alpha = 0.2$) | | BNL | OTRSH | reference |
	($\gamma = 0.25$ Bohr^{-1})	($\gamma = 0.40$ Bohr^{-1})			
Benzene	9.40	9.48	9.42	9.40	9.45
Naphthalene	7.85	8.27	8.19	8.09	8.25
Anthracene	7.13	7.49	7.40	7.31	7.48
Tetracene	6.65	6.96	6.86	6.75	6.96
Pentacene	6.31	6.59	6.42	6.31	6.58
Hexacene	6.05	6.30	6.07	6.01	6.32
MAD	0.27	0.02	0.11	0.19	

Table S7: Ionization potential energies calculated with G_0W_0. All energies are in units of eV and γ values are in given in Bohr^{-1}.

	G_0W_0 @						CCSD(T)
	PBE	PBE0	BHLYP	RSH ($\alpha = 0.2$)		OTRSH	reference
				($\gamma = 0.25$)	($\gamma = 0.40$)		
Benzene	9.14	9.35	9.52	9.54	9.77	9.54	9.45
Naphthalene	7.96	8.17	8.33	8.45	8.60	8.35	8.25
Anthracene	7.21	7.41	7.57	7.68	7.83	7.57	7.48
Tetracene	6.68	6.88	7.04	7.16	7.31	7.03	6.96
Pentacene	6.32	6.51	6.67	6.79	6.94	6.65	6.58
Hexacene	6.04	6.23	6.38	6.51	6.66	6.37	6.32
MAD	0.28	0.08	0.08	0.18	0.35	0.08	

Table S8: Ionization potential energies calculated with evGW_0 and evGW (in eV).

	evGW_0 @			evGW @			CCSD(T)
	PBE	PBE0	BHLYP	PBE	PBE0	BHLYP	reference
Benzene	9.36	9.52	9.56	9.55	9.55	9.61	9.45
Naphthalene	8.15	8.24	8.37	8.32	8.33	8.41	8.25
Anthracene	7.37	7.47	7.60	7.53	7.55	7.63	7.48
Tetracene	6.84	6.92	7.05	6.99	7.02	7.10	6.96
Pentacene	6.46	6.56	6.70	6.61	6.64	6.72	6.58
Hexacene	6.19	6.28	6.39	6.32	6.35	6.44	6.32
MAD	0.11	0.03	0.10	0.05	0.07	0.15	

Table S9: Electron affinity energies calculated with DFT (in eV).

	DFT				CCSD(T)
	RSH ($\alpha = 0.2$)		BNL	OTRSH	reference
	($\gamma = 0.25$ Bohr^{-1})	($\gamma = 0.40$ Bohr^{-1})			
Benzene	-1.54	-2.16	-1.66	-1.54	-1.53
Naphthalene	-0.90	-1.17	-0.49	-0.36	-0.48
Anthracene	-0.16	-0.40	0.33	0.44	0.28
Tetracene	0.36	0.16	0.90	1.03	0.82
Pentacene	0.73	0.57	1.33	1.41	1.21
Hexacene	1.02	0.88	1.69	1.77	1.47
MAD	0.38	0.65	0.10	0.17	

Table S10: Electron affinity energies calculated with G_0W_0 (in eV). All energies are in units of eV and γ values are in given in Bohr^{-1}.

	G_0W_0 @						CCSD(T)
	PBE	PBE0	BHLYP	RSH ($\alpha = 0.2$)		OTRSH	reference
				($\gamma = 0.25$)	($\gamma = 0.40$)		
Benzene	-0.96	-1.16	-1.23	-1.39	-1.28	-1.39	-1.53
Naphthalene	0.21	-0.01	-0.12	-0.19	-0.17	-0.28	-0.48
Anthracene	1.02	0.80	0.69	0.61	0.64	0.56	0.28
Tetracene	1.58	1.37	1.26	1.18	1.21	1.15	0.82
Pentacene	1.99	1.78	1.67	1.59	1.62	1.58	1.21
Hexacene	2.30	2.09	1.98	1.89	1.92	1.90	1.47
MAD	0.73	0.52	0.41	0.32	0.36	0.29	

Table S11: Electron affinity energies calculated with evGW_0 and evGW (in eV).

	evGW_0 @			evGW @			CCSD(T)
	PBE	PBE0	BHLYP	PBE	PBE0	BHLYP	reference
Benzene	-1.13	-1.34	-1.25	-1.37	-1.37	-1.30	-1.53
Naphthalene	0.05	-0.09	-0.14	-0.17	-0.21	-0.19	-0.48
Anthracene	0.87	0.73	0.67	0.67	0.62	0.63	0.28
Tetracene	1.44	1.30	1.24	1.25	1.20	1.21	0.82
Pentacene	1.86	1.72	1.66	1.68	1.62	1.63	1.21
Hexacene	2.15	2.03	1.97	1.99	1.94	1.94	1.47
MAD	0.58	0.43	0.40	0.38	0.34	0.36	

3 OTRSH convergence

Here we show convergence studies on the charged excitations calculated within OTRSH. In Tables S12 and S13 we show IP and EA energies calculated within OTRSH using basis sets of increasing size. Note that for the larger acenes, some entries are missing in the Tables since the calculations are computationally unfeasible. Hence, we resort to similar extrapolations as in Section 1.2. Yellow cells imply a linear-extrapolation based on the trend on that column (from smaller acenes), while blue cells indicates that the Swartz formula, Eq. 2, was used to obtain the ∞Z results. By comparing to this CBS reference, we can estimate that using the aTZ basis the IP/EA energies are converged within better than 0.01/0.02 eV.

Table S12: IP energy calculated with the OTRSH, $\mathrm{IP^{OTRSH}}$, using different basis sets. The results within aDZ basis and energy difference between IPs calculated within different basis sets are shown (read text).

	aDZ	aTZ−aDZ	aQZ−aTZ	∞Z−aQZ
		$\mathrm{IP^{OTRSH}}$ (eV)		
Benzene	9.38	0.021	0.003	0.000
Naphthalene	8.07	0.015	0.003	0.002
Anthracene	7.29	0.013	0.004	0.002
Tetracene	6.74	0.012	0.004	0.002
Pentacene	6.33	0.010	0.004	0.003
Hexacene	6.05	0.009	0.004	0.003

Table S13: EA energy calculated with the OTRSH, $\mathrm{EA^{OTRSH}}$ using different basis sets. The same convention as in Table. S12 is used.

	aDZ	aTZ−aDZ	aQZ−aTZ	∞Z−aQZ
		$\mathrm{EA^{OTRSH}}$ (eV)		
Benzene		-1.57	0.029	0.011	0.008
Naphthalene		-0.38	0.020	0.004	0.002
Anthracene		0.43	0.017	0.004	0.002
Tetracene		1.01	0.015	0.004	0.002
Pentacene		1.45	0.014	0.004	0.002
Hexacene		1.75	0.013	0.004	0.002

4 Geometries

Here we provide the relaxed geometries of the oligoacenes used in this work. The structures were relaxed with Q-Chem 4.2 software package.[S5] and using the B3LYP exchange-correlation functional.

Table S14: Atomic positions of benzene in units of Å

C	0.000000	1.392033	0.000000	C	0.000000	-1.392033	0.000000
C	1.205536	0.696016	0.000000	C	-1.205536	-0.696016	0.000000
C	-1.205536	0.696016	0.000000	C	1.205536	-0.696016	0.000000
H	0.000000	2.475387	0.000000	H	0.000000	-2.475387	0.000000
H	2.143748	1.237694	0.000000	H	-2.143748	-1.237694	0.000000
H	-2.143748	1.237694	0.000000	H	2.143748	-1.237694	0.000000

Table S15: Atomic positions of naphthalene in units of Å

C	0.000000	0.000000	0.714895	C	0.000000	0.000000	-0.714895
C	0.000000	1.241902	1.398324	C	0.000000	-1.241902	-1.398324
C	0.000000	-1.241902	1.398324	C	0.000000	1.241902	-1.398324
C	0.000000	2.426173	0.706201	C	0.000000	-2.426173	-0.706201
C	0.000000	-2.426173	0.706201	C	0.000000	2.426173	-0.706201
H	0.000000	1.241617	2.482398	H	0.000000	-1.241617	-2.482398
H	0.000000	-1.241617	2.482398	H	0.000000	1.241617	-2.482398
H	0.000000	3.367735	1.241610	H	0.000000	-3.367735	-1.241610
H	0.000000	-3.367735	1.241610	H	0.000000	3.367735	-1.241610

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Table S16: Atomic positions of anthracene in units of Å

C	-1.220168	0.720757	0.000000	C	-2.473246	1.402706	0.000000
C	-3.649834	0.710903	0.000000	C	-3.649845	-0.710930	0.000000
C	-2.473239	-1.402727	0.000000	C	-1.220175	-0.720752	0.000000
C	1.220183	0.720735	0.000000	C	0.000000	1.399698	0.000000
C	-0.000017	-1.399715	0.000000	C	1.220174	-0.720777	0.000000
C	3.649856	0.710946	0.000000	C	2.473215	1.402709	0.000000
C	2.473241	-1.402683	0.000000	C	3.649853	-0.710861	0.000000
H	-2.473723	2.486682	0.000000	H	-4.593210	1.242978	0.000000
H	-4.593179	-1.243070	0.000000	H	-2.473666	-2.486698	0.000000
H	0.000000	2.484534	0.000000	H	-0.000004	-2.484549	0.000000
H	2.473654	2.486686	0.000000	H	2.473803	-2.486659	0.000000
H	4.593177	1.243106	0.000000	H	4.593153	-1.243067	0.000000

Table S17: Atomic positions of tetracene in units of Å

C	-4.874668	-0.713102	0.000005	C	-3.701529	-1.404960	0.000083
C	-2.443371	-0.724213	0.000013	C	-1.231979	-1.402550	-0.000051
C	-0.000017	-0.724341	-0.000063	C	-1.232009	1.402428	0.000069
C	-2.443357	0.723928	-0.000003	C	-3.701432	1.404961	-0.000059
C	-4.874677	0.713280	-0.000031	C	0.000017	0.724342	0.000062
C	1.232009	-1.402428	-0.000070	C	1.231979	1.402550	0.000050
C	2.443357	-0.723928	0.000003	C	3.701432	-1.404961	0.000059
C	2.443371	0.724213	-0.000013	C	4.874677	-0.713280	0.000032
C	3.701529	1.404960	-0.000082	C	4.874668	0.713102	-0.000004
H	5.818867	1.243634	-0.000043	H	-5.818866	-1.243635	0.000044
H	-3.702867	-2.488887	0.000052	H	-1.232443	-2.487235	-0.000121
H	-1.232810	2.487118	0.000055	H	-3.702217	2.488876	-0.000024
H	-5.818840	1.243876	-0.000049	H	1.232810	-2.487118	-0.000056
H	3.702868	2.488887	-0.000052	H	5.818839	-1.243877	0.000050
H	1.232443	2.487235	0.000121	H	3.702217	-2.488876	0.000024

Table S18: Atomic positions of pentacene in units of Å

C	-0.513800	-6.120281	0.002055	C	0.513800	6.120281	-0.002055
C	-1.243594	-4.972398	0.000101	C	1.243594	4.972398	-0.000101
C	-0.604869	-3.689503	0.001050	C	0.604869	3.689503	-0.001050
C	-1.322430	-2.505713	-0.001240	C	1.322430	2.505713	0.001240
C	-0.686015	-1.245946	-0.000584	C	0.686015	1.245946	0.000584
C	-1.403859	-0.046091	-0.002964	C	1.403859	0.046091	0.002964
C	0.766464	-1.198497	0.002526	C	-0.766464	1.198497	-0.002526
C	1.484277	-2.413569	0.005091	C	-1.484277	2.413569	-0.005091
C	0.846020	-3.642063	0.004303	C	-0.846020	3.642063	-0.004303
C	1.567650	-4.880066	0.006537	C	-1.567650	4.880066	-0.006537
C	0.914860	-6.073482	0.005228	C	-0.914860	6.073482	-0.005228
H	-1.011577	-7.082146	0.001114	H	1.011577	7.082146	-0.001114
H	-2.327049	-5.008369	-0.002191	H	2.327049	5.008369	0.002191
H	-2.406635	-2.542013	-0.003610	H	2.406635	2.542013	0.003610
H	-2.487971	-0.081873	-0.005427	H	2.487971	0.081873	0.005427
H	2.568507	-2.378555	0.007504	H	-2.568507	2.378555	-0.007504
H	2.651117	-4.844869	0.009345	H	-2.651117	4.844869	-0.009345
H	1.474859	-7.000644	0.006729	H	-1.474859	7.000644	-0.006729

Table S19: Atomic positions of hexacene in units of Å

C	-3.688435	-1.405128	-0.000486	C	-0.000002	-0.728637	-0.000019
C	-1.228975	-1.405931	-0.000313	C	-6.155456	1.407090	0.000003
C	-7.325641	0.715488	0.000437	C	-7.325639	-0.715467	0.000501
C	-6.155475	-1.407091	0.000102	C	-4.892783	-0.726914	-0.000258
C	-4.892777	0.726896	-0.000288	C	-3.688428	1.405105	-0.000468
C	-2.447159	0.728043	-0.000437	C	-2.447166	-0.728067	-0.000475
C	-1.228981	1.405927	-0.000203	C	6.155455	-1.407094	-0.000024
C	7.325641	-0.715492	-0.000471	C	7.325640	0.715461	-0.000554
C	6.155473	1.407084	-0.000117	C	4.892782	0.726909	0.000265
C	4.892777	-0.726900	0.000289	C	3.688425	-1.405109	0.000470
C	2.447162	-0.728040	0.000452	C	2.447168	0.728068	0.000499
C	1.228977	-1.405915	0.000214	C	3.688434	1.405128	0.000513
C	0.000002	0.728649	0.000041	C	1.228977	1.405937	0.000342
H	3.689485	2.489891	0.000753	H	1.229350	2.490580	0.000426
H	8.270848	1.244307	-0.000923	H	6.156431	2.491100	-0.000151
H	-3.689463	-2.489891	-0.000734	H	-1.229338	-2.490574	-0.000387
H	6.156397	-2.491111	0.000124	H	8.270834	-1.244354	-0.000804
H	3.689471	-2.489873	0.000728	H	1.229378	-2.490559	0.000257
H	-6.156399	2.491108	-0.000137	H	-8.270840	1.244339	0.000765
H	-3.689476	2.489871	-0.000713	H	-1.229379	2.490571	-0.000244
H	-6.156426	-2.491106	0.000120	H	-8.270848	-1.244312	0.000832