SUPPORTING INFORMATION: Influence of the crystal packing in singlet fission: one step beyond the gas phase approximation

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1 Analysis of the AC tetracene dimer with implicit environment under the C-PCM approach

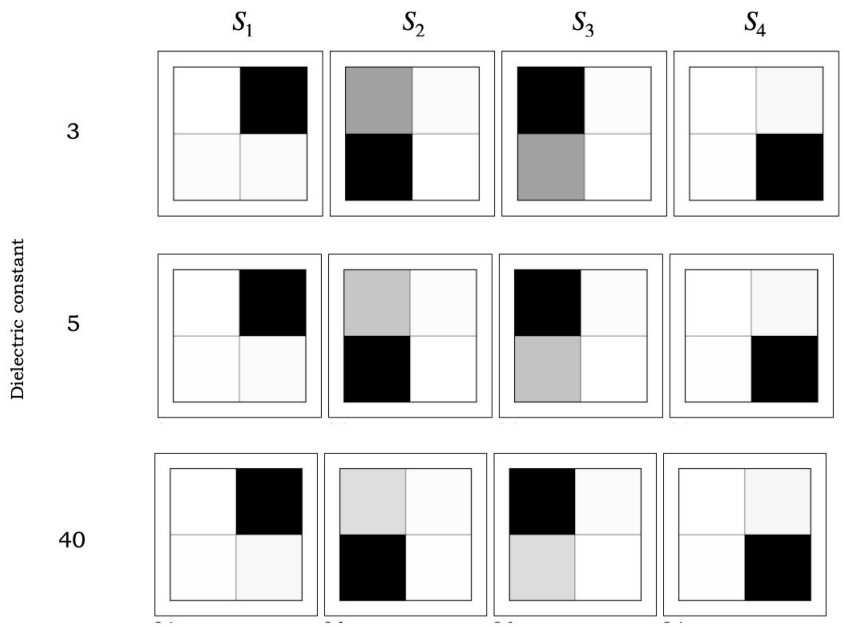

Figure S1: *Electron-hole* correlation plots of the four lowest singlet excited states of the AC tetracene dimer calculated at the CAM-B3LYP/ANO-S-DZV level of theory with implicit consideration of the environment using the conductor-like polarizable continuum model. Three different dielectric constant values were tested.

Table S1: Exciton descriptors of the AC tetracene dimer calculated at the CAM-B3LYP/ANO-S-DZV level of theory with implicit consideration of the environment using the conductor-like polarizable continuum model. Different values of dielectric constant were tested.

Dielectric constant	State	$\Delta E(eV)$	f	ω_{CT}	d_{exc}(Å)	$d_{h\rightarrow e}$(Å)	R_{eh}
3	S_1	2.832	0.131	0.041	4.656	0.168	0.087
	S_2	2.905	0.193	0.310	5.343	1.446	0.087
	S_3	3.119	0.091	0.670	6.048	3.210	0.066
	S_4	3.382	0.007	0.932	6.546	4.528	0.045
5	S_1	2.830	0.132	0.046	4.668	0.191	0.083
	S_2	2.915	0.203	0.246	5.204	1.129	0.088
	S_3	3.146	0.078	0.733	6.166	3.516	0.063
	S_4	3.347	0.008	0.932	6.546	4.527	0.044
40	S_1	2.828	0.132	0.054	4.689	0.230	0.079
	S_2	2.924	0.224	0.182	5.064	0.817	0.089
	S_3	3.184	0.055	0.793	6.280	3.806	0.058
	S_4	3.301	0.011	0.927	6.535	4.498	0.043

2 NOCI coefficients for dimer AB of tetracene in gas phase

Table S2: NOCI coefficients of the MEBFs for each of the states in the AB tetracene dimer in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0163879947	0.0326184562	-0.0017143516	0.0186147718	-0.0116895214
$\Psi_{S_0S_1}$	**-0.6886361256**	**-0.6812479975**	0.0051490887	0.1646688122	0.1844254748
$\Psi_{S_1S_0}$	**0.6981237177**	**-0.6720588219**	-0.0053941845	-0.1103149489	0.2209966565
$\Psi_{D_0^+D_0^-}$	-0.0183301187	0.0001893425	-0.0332884331	**-0.7807105286**	**-0.5800879843**
$\Psi_{D_0^-D_0^+}$	0.1287149044	-0.1923192327	0.0327418027	**0.5959303929**	**-0.7684664354**
$\Psi_{^1TT}$	-0.1272566329	-0.1924907077	**-0.9984300281**	-0.0532340344	0.0076356900

3 NOCI coefficients for dimer AB of tetracene within environment

Table S3: NOCI coefficients of the MEBFs for each of the states in the AB tetracene dimer within the embedded cluster approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0207713963	0.0015764954	-0.0000151766	0.0018742604	-0.0008645368
$\Psi_{S_0S_1}$	**-0.7374600180**	**0.6745428844**	0.0000488845	-0.0102349033	-0.0301137224
$\Psi_{S_1S_0}$	**0.6743392994**	**0.7380078848**	-0.0000308424	0.0210679303	-0.0043377444
$\Psi_{D_0^+D_0^-}$	0.0195308205	0.0069816432	0.0015979660	**-0.9994027226**	-0.0277567804
$\Psi_{D_0^-D_0^+}$	0.0165864516	-0.0207389411	-0.0008885431	0.0279185778	**-0.9992637640**
$\Psi_{^1TT}$	-0.0000203579	0.0000579268	**-0.9999968791**	-0.0022977309	0.0012605533

4 NOCI coefficients for dimer AC of tetracene in gas phase

Table S4: NOCI coefficients of the MEBFs for each of the states in the AC tetracene dimer in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.1142570343	-0.1619990402	-0.0105682548	0.1773237073	-0.0326870036
$\Psi_{S_0S_1}$	**0.9776315772**	0.1925162022	0.0156587538	-0.0717921849	-0.0371242419
$\Psi_{S_1S_0}$	0.1127826315	0**.7292796938**	-0.0035909317	-0.4661727606	0.4881599628
$\Psi_{D_0^+D_0^-}$	-0.1344991129	0.6301805346	0.0065742959	**0.8502714014**	0.0541986962
$\Psi_{D_0^-D_0^+}$	-0.0155512693	0.0853111102	0.0044843290	0.1028896737	**0.8775119150**
$\Psi_{^1TT}$	-0.0008624642	-0.0002790514	**-1.0001993844**	-0.0052073806	0.0022836208

5 NOCI coefficients for dimer AC of tetracene within environment

Table S5: NOCI coefficients of the MEBFs for each of the states in the AC tetracene dimer within the embedded cluster approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.1214496443	0.2737342658	-0.0076988190	0.1626828163	-0.4028476716
$\Psi_{S_0S_1}$	**-0.9700027448**	-0.2242079872	-0.0118892816	-0.4632671256	0.0220587496
$\Psi_{S_1S_0}$	0.2227403069	**-0.7697610529**	-0.0105456983	**0.8566024814**	-0.3679058980
$\Psi_{D_0^+D_0^-}$	-0.0006317668	-0.5159896609	-0.0058505293	-0.1379450703	**-0.8341319151**
$\Psi_{^1TT}$	0.0009090031	0.0045715229	**0.9999060427**	-0.0000951191	-0.0050949128

6 NOCI coefficients for dimer BC of tetracene in gas phase

Table S6: NOCI coefficients of the MEBFs for each of the states in the BC tetracene dimer in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0948382203	0.1144850107	0.0103434661	-0.0121419427	-0.0154864420
$\Psi_{S_0S_1}$	**-0.7445303757**	0.6672794993	0.0139278977	0.0108810358	0.0078507072
$\Psi_{S_1S_0}$	-0.6617600665	**-0.7349053925**	0.0147873633	-0.0023559670	-0.0192926440
$\Psi_{D_0^+D_0^-}$	0.0017872204	-0.0177822683	-0.0087268439	**-0.9965219340**	-0.0815798208
$\Psi_{D_0^-D_0^+}$	0.0044995716	0.0267001167	-0.0007002834	-0.0763797372	**0.9969768806**
$\Psi_{^1TT}$	-0.0014904133	-0.0012590100	**1.0003418214**	0.0001167292	0.0002532096

7 NOCI coefficients for dimer BC of tetracene within environment

Table S7: NOCI coefficients of the MEBFs for each of the states in the BC tetracene dimer within the embedded cluster approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0276858840	0.0008523775	0.0296876879	-0.0094299895	0.0396862053
$\Psi_{S_0S_1}$	**0.9983813709**	0.0026697629	-0.1241579979	-0.0119924253	0.1330118814
$\Psi_{S_1S_0}$	0.0056155406	**-1.0221199346**	0.1093015989	-0.0432489053	-0.5970448638
$\Psi_{D_0^+D_0^-}$	-0.0012001336	0.1270197565	-0.0799320488	**-0.9984832904**	-0.1597561767
$\Psi_{D_0^-D_0^+}$	0.0153592053	-0.0360086821	0.0595511914	-0.0173866486	**-1.2070663758**
$\Psi_{^1TT}$	-0.0040124666	0.0050369378	**1.0140154532**	0.0004415150	0.0068675524

8 AIFDEM eigevectors in the exciton-site basis for the I.a-I.b of α in the gas phase

Table S8: AIFDEM coefficients of the MEBFs for each of the states in the I.a-I.b DPBF dimer of α in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0277737	0.0028854	-0.0007588	-0.0064482	0.0215616
$\Psi_{S_0S_1}$	**-0.7324108**	**0.7077950**	0.0034109	-0.0623480	0.0082529
$\Psi_{S_1S_0}$	**0.7093863**	**0.7308431**	-0.0033406	-0.0478294	-0.0395819
$\Psi_{D_0^+D_0^-}$	0.0278592	-0.0633481	-0.0290414	**-0.8722924**	0.4831785
$\Psi_{D_0^-D_0^+}$	0.0250671	0.0636147	-0.0289742	0.4811448	**0.8734818**
$\Psi_{^1TT}$	0.0064934	0.0000345	**1.0940155**	-0.0108550	0.0374750

9 AIFDEM eigevectors in the exciton-site basis for the I.a-I.b in α within environment

Table S9: NOCI coefficients of the MEBFs for each of the states in the I.a-I.b DPBF dimer of α within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.0000191	-0.0000144	-0.0000009	-0.0000191	-0.0000144
$\Psi_{S_0S_1}$	**-0.9997001**	0.0018118	-0.0246769	0.0000350	-0.0000133
$\Psi_{S_1S_0}$	0.0008422	**0.9992429**	0.0389297	-0.0000129	0.0000294
$\Psi_{D_0^+D_0^-}$	0.0000350	-0.0000133	0.0000003	**-0.9997001**	0.0018118
$\Psi_{D_0^-D_0^+}$	-0.0000129	0.0000294	0.0000007	0.0008422	**0.9992429**
$\Psi_{^1TT}$	-0.0223357	-0.0394457	**1.0929689**	-0.0223357	-0.0394457

10 AIFDEM eigevectors in the exciton-site basis for the I.a-I.b in β in gas phase

Table S10: AIFDEM coefficients of the MEBFs for each of the states in the I.a-I.b DPBF dimer of β in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.0418105	0.0086049	-0.0008397	0.0054538	0.0205476
$\Psi_{S_0S_1}$	**0.7382234**	**0.7010222**	0.0034165	-0.0491583	0.0436482
$\Psi_{S_1S_0}$	**-0.7021828**	**0.7365430**	-0.0035112	-0.0666754	-0.008790
$\Psi_{D_0^+D_0^-}$	-0.0299693	-0.0653714	-0.0296590	-0.4611530	**0.8839012**
$\Psi_{D_0^-D_0^+}$	-0.0279021	0.0680036	-0.0292485	**0.8825973**	0.4634259
$\Psi_{^1TT}$	-0.0067833	0.0002505	**1.0924029**	0.0115278	0.0379143

11 AIFDEM eigevectors in the exciton-site basis for the I.a-I.b in β within environment

Table S11: NOCI coefficients of the MEBFs for each of the states in the I.a-I.b DPBF dimer of β within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.0167263	0.0164963	-0.0007205	0.0050219	0.0219513
$\Psi_{S_0S_1}$	**0.7347262**	**0.7052121**	0.0029714	-0.0497610	0.0411570
$\Psi_{S_1S_0}$	**-0.7070430**	**0.7324009**	-0.0032869	-0.0662809	-0.0074880
$\Psi_{D_0^+D_0^-}$	-0.0278757	-0.0648880	-0.0296331	-0.4592395	**0.8849997**
$\Psi_{D_0^-D_0^+}$	-0.0266463	0.0683569	-0.0292400	**0.8835906**	0.4615508
$\Psi_{^1TT}$	-0.0061651	0.0004055	**1.0924061**	0.0116089	0.0378994

12 AIFDEM eigevectors in the exciton-site basis for the I-II in α in the gas phase

Table S12: AIFDEM coefficients of the MEBFs for each of the states in the I-II DPBF dimer of α in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.0039296	-0.002	-0.0000002	-0.0000617	0.0003244
$\Psi_{S_0S_1}$	0.5240654	**0.8757561**	0.0000178	0.0010823	-0.0021444
$\Psi_{S_1S_0}$	**0.8757588**	-0.5240718	-0.0000449	-0.0021920	0.0011817
$\Psi_{D_0^+D_0^-}$	0.0000893	0.0024594	-0.0002189	-0.0000362	**0.9999969**
$\Psi_{D_0^-D_0^+}$	0.0013220	-0.0020684	0.0038038	**0.9999898**	0.0000421
$\Psi_{^1TT}$	0.0000263	-0.0000323	**1.0947486**	-0.0040753	0.0002446

13 AIFDEM eigevectors in the exciton-site basis for the I-II in α within environment

Table S13: NOCI coefficients of the MEBFs for each of the states in the I-II DPBF dimer of α within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Phi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	-0.0000381	-0.0000144	-0.0000009	-0.0000001	-0.000001
$\Psi_{S_0S_1}$	0.0008422	**0.9992429**	-0.0246769	-0.0000002	-0.0000003
$\Psi_{S_1S_0}$	**-0.99950021**	0.0008221	0.0389297	0.0000001	-0.0000002
$\Psi_{D_0^+D_0^-}$	0.0000350	-0.0000133	0.0000003	0.0000001	**0.9999987**
$\Psi_{D_0^-D_0^+}$	-0.0000129	0.0000294	0.0000007	**0.9999991**	-0.0000002
$\Psi_{^1TT}$	-0.0223357	-0.0394457	**1.0929689**	-0.0001920	-0.0019023

14 AIFDEM eigevectors in the exciton-site basis for the I-II in β in the gas phase

Table S14: AIFDEM coefficients of the MEBFs for each of the states in the I-II DPBF dimer of β in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0258270	0.0092295	0.0000018	0.0001313	0.0003338
$\Psi_{S_0S_1}$	**0.7974221**	0.6361385	0.0000625	0.0024779	0.0013158
$\Psi_{S_1S_0}$	-0.6361485	**0.7978020**	0.0000293	0.0012771	0.0025338
$\Psi_{D_0^+D_0^-}$	-0.0011368	-0.0025518	0.0044709	**0.9999861**	-0.0000535
$\Psi_{D_0^-D_0^+}$	-0.0005439	0.0028084	-0.0003806	-0.0000452	**-0.9999958**
$\Psi_{^1TT}$	-0.0000277	-0.0000532	**1.0931556**	-0.0047956	-0.0004188

15 AIFDEM eigevectors in the exciton-site basis for the I-II in β within environment

Table S15: NOCI coefficients of the MEBFs for each of the states in the I-II DPBF dimer of β within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{^1TT}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$
$\Psi_{S_0S_0}$	0.0000047	-0.0000107	-0.0000000	-0.0000003	-0.0000000
$\Psi_{S_0S_1}$	**1.0206298**	0.0000314	-0.000000	-0.0000002	-0.0000002
$\Psi_{S_1S_0}$	-0.0000315	**1.0206264**	-0.0000000	-0.0000002	-0.0000002
$\Psi_{D_0^+D_0^-}$	-0.0000290	-0.0000496	0.0012316	0.0000001	**0.9999992**
$\Psi_{D_0^-D_0^+}$	-0.0000348	1.0000000	0.000218	**1.0000000**	-0.0000003
$\Psi_{^1TT}$	-0.0000000	-0.0002087	**1.0934487**	-0.0002087	-0.0014092

16 AIFDEM eigevectors in the exciton-site basis for the trimer in α in gas phase

Table S16: AIFDEM coefficients of the MEBFs for each of the states in the DPBF trimer of α in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{S[3]}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$	$\Phi_{CT[3]}$
$\Psi_{S_0S_0S_0}$	-0.0038108	0.0025470	-0.0022037	0.0000000	0.0000042	-0.0000951
$\Psi_{S_1S_0S_0}$	**0.3325867**	**-0.6800892**	**-0.6844498**	0.0000000	-0.0000000	0.0000000
$\Psi_{S_0S_1S_0}$	**0.6875237**	**0.6742966**	**-0.3358551**	-0.0000000	-0.0000478	0.0000282
$\Psi_{S_0S_0S_1}$	**0.6764227**	**-0.3519160**	**0.6783672**	0.0000000	0.0000347	-0.0000531
$\Psi_{D_0^+D_0^-S_0}$	-0.0000006	0.0000034	0.0000013	0.0000000	0.0000000 -	-0.0000414
$\Psi_{D_0^+S_0D_0^-}$	-0.0000856	-0.0009760	-0.0020886	-0.0000499	-0.0000000	-0.0000000
$\Psi_{D_0^-D_0^+S_0}$	-0.0000032	-0.0000035	-0.0000006	**1.0000000**	-0.0000499	0.0000000
$\Psi_{S_0D_0^+D_0^-}$	0.0007365	0.0018191	-0.0014328	0.0000000	**1.0000000**	-0.0000000
$\Psi_{D_0^-S_0D_0^+}$	0.0011920	-0.0000069	0.0023457	-0.0000000	0.0000000	0.0000000
$\Psi_{S_0D_0^-D_0^+}$	0.0006568	-0.0015543	0.0018149	-0.0000414	-0.0000000	**1.0000000**
$\Psi_{^1(TTS_0)}$	0.0293356	0.0253935	-0.0128473	-0.0000000	0.0000000	-0.0000000
$\Psi_{^1(TS_0T)}$	0.0000229	-0.0000012	0.0000356	-0.0000000	0.0000000	-0.0000000
$\Psi_{^1(S_0TT)}$	0.0000144	0.0000295	-0.0000209	0.0000000	-0.0000744	-0.0000042

17 AIFDEM eigevectors in the exciton-site basis for the trimer in α within environment

Table S17: NOCI coefficients of the MEBFs for each of the states in the DPBF trimer of α within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{S[3]}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$	$\Phi_{CT[3]}$
$\Psi_{S_0S_0S_0}$	-0.0000000	0.0000000	-0.0000000	0.0000000	-0.0001001	0.0000003
$\Psi_{S_1S_0S_0}$	**0.9604347**	-0.0000000	-0.0000000	0.0000000	0.0000000	-0.0000000
$\Psi_{S_0S_1S_0}$	-0.0000000	**0.9608577**	0.0000000	0.0000000	0.0000287	0.0000552
$\Psi_{S_0S_0S_1}$	-0.0000000	0.0000000	**0.9608583**	-0.0000000	0.0000529	0.0000372
$\Psi_{D_0^+D_0^-S_0}$	0.0000000	0.0000000	-0.0000000	0.0000000	-0.0000458	-0.0000000
$\Psi_{D_0^+S_0D_0^-}$	-0.0000533	0.0000000	0.0000283	-0.0000000	0.0000000	-0.0000000
$\Psi_{D_0^-D_0^+S_0}$	0.0000000	0.0000000	-0.0000000	**1.0000000**	-0.0000000	-0.0000578
$\Psi_{S_0D_0^+D_0^-}$	-0.0000000	0.0000552	0.0000372	-0.0000578	-0.0000000	**1.0000000**
$\Psi_{D_0^-S_0D_0^+}$	0.0000373	0.0000000	-0.0000567	-0.0000462	-0.0000000	0.0000000
$\Psi_{S_0D_0^-D_0^+}$	0.0000000	0.0000287	0.0000529	-0.0000000	**1.0000000**	-0.0000000
$\Psi_{^1(TTS_0)}$	0.0000000	-0.0000000	-0.0000000	0.0000000	-0.0000000	-0.0000000
$\Psi_{^1(TS_0T)}$	0.0000000	-0.0000000	-0.0000000	0.0000000	-0.0000000	-0.0000000
$\Psi_{^1(S_0TT)}$	-0.0000000	0.0000000	0.0000000	-0.0000000	-0.000013	0.0000861

18 AIFDEM eigevectors in the exciton-site basis for the trimer in β in gas phase

Table S18: AIFDEM coefficients of the MEBFs for each of the states in the DPBF trimer of β in the gas phase. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{S[3]}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$	$\Phi_{CT[3]}$
$\Psi_{S_0S_0S_0}$	0.0143643	-0.0062035	-0.0117932	0.0002852	-0.0000001	0.0000000
$\Psi_{S_1S_0S_0}$	**-0.6578650**	**-0.3454719**	**0.6991897**	0.0014282	-0.0000007	-0.0000004
$\Psi_{S_0S_1S_0}$	**0.4226623**	**0.6110173**	**0.6994453**	0.0023250	0.0000015	-0.0000009
$\Psi_{S_0S_0S_1}$	**0.6554500**	**-0.7406173**	**0.2510053**	-0.0000766	-0.0000007	0.0000021
$\Psi_{D_0^+D_0^-S_0}$	0.0012150	0.0000714	-0.0027949	0.0002872	-0.0000000	0.0008446
$\Psi_{D_0^+S_0D_0^-}$	0.0007670	0.0019238	-0.0021050	-0.0000220	0.0007789	0.0000000
$\Psi_{D_0^-D_0^+S_0}$	-0.0000011	0.0009655	0.0025569	**-0.9999957**	0.0009816	-0.0000001
$\Psi_{S_0D_0^+D_0^-}$	-0.0000011	-0.0000039	-0.0000012	0.0009320	0**.9999993**	0.0000000
$\Psi_{D_0^-S_0D_0^+}$	0.0007699	-0.0022476	0.0014872	0.0000429	-0.0000000	0.0010527
$\Psi_{S_0D_0^-D_0^+}$	-0.0000029	0.0000040	0.0000011	-0.0000003	-0.0000000	**0.9999992**
$\Psi_{^1(TTS_0)}$	0.0000315	0.0000025	-0.0000566	-0.0003826	0.0000000	0.0000020
$\Psi_{^1(TS_0T)}$	-0.0000194	0.0000496	-0.0000290	0.0000002	0.0000004	-0.0000033
$\Psi_{^1(S_0TT)}$	-0.0011484	-0.0027377	-0.0021242	0.0000023	-0.00000004	0.0000000

19 AIFDEM eigevectors in the exciton-site basis for the trimer in β within environment

Table S19: NOCI coefficients of the MEBFs for each of the states in the DPBF trimer of β within the QM/MM approach. The largest value(s) is(are) indicated in bold.

MEBF	$\Phi_{S[1]}$	$\Phi_{S[2]}$	$\Psi_{S[3]}$	$\Phi_{CT[1]}$	$\Phi_{CT[2]}$	$\Phi_{CT[3]}$
$\Psi_{S_0S_0S_0}$	-0.0000105	0.0000015	-0.0000307	-0.0000000	-0.0000000	-0.0000003
$\Psi_{S_1S_0S_0}$	0.0000477	0.0000384	**1.0204279**	-0.0000004	-0.0000000	-0.0000002
$\Psi_{S_0S_1S_0}$	**1.0206066**	-0.0000000	-0.0000477	-0.0000000	0.0000000	-0.0000002
$\Psi_{S_0S_0S_1}$	-0.0000000	**1.0206080**	-0.0000384	0.0000002	0.0000000	-0.0000000
$\Psi_{D_0^+D_0^-S_0}$	0.0000504	0.0000000	0.0000295	0.0000003	-0.0009080	-0.0000001
$\Psi_{D_0^+S_0D_0^-}$	-0.0000000	0.0000252	-0.0000333	-0.0000006	0.0000000	-0.0000003
$\Psi_{D_0^-D_0^+S_0}$	0.0000345	-0.0000000	0.0000251	-0.0000000	-0.0000000	**-0.9999998**
$\Psi_{S_0D_0^+D_0^-}$	0.0000000	0.0000000	-0.0000000	-0.0000000	0.0000000	0.0004648
$\Psi_{D_0^-S_0D_0^+}$	-0.0000000	0.0000306	-0.0000507	**0.9999982**	0.0005785	-0.0000000
$\Psi_{S_0D_0^-D_0^+}$	-0.0000000	-0.0000000	0.0000000	-0.0006079	**0.9999995**	-0.0000000
$\Psi_{^1(TTS_0)}$	-0.0000000	0.0000000	-0.0000000	-0.0000000	0.0000001	-0.0002604
$\Psi_{^1(TS_0T)}$	-0.0000000	0.0000000	-0.0000000	-0.0020380	-0.0000001	-0.0000000
$\Psi_{^1(S_0TT)}$	-0.0000001	-0.0000000	0.0000000	0.0000000	0.0000000	-0.0000000

20 Cartesian coordinates of monomer A (Tetracene)

```
C -3.488181 1.248517 29.291033
C -2.845556 2.130006 28.468334
C -2.789402 1.869723 27.022786
C -2.157667 2.718182 26.189052
C -2.072096 2.469712 24.780287
C -1.463320 3.306282 23.888928
C -3.325788 0.672418 26.528676
C -4.027745 -0.22365 27.409001
C -4.087748 0.031585 28.792019
H -3.509941 1.482638 30.284157
H -2.432966 3.040049 28.800601
H -1.722242 3.645335 26.544615
H -1.044095 4.238807 24.251847
H -4.363485 -1.11918 26.998265
H -4.524619 -0.65239 29.425902
C -2.669781 1.246873 24.262882
C -1.416089 3.044167 22.514493
C -3.278556 0.410303 25.154242
C -2.584209 0.998404 22.854117
C -1.952474 1.846863 22.020383
C -0.714131 3.940244 21.634168
H -3.697781 -0.52222 24.791322
H -3.019635 0.071251 22.498554
C -1.896320 1.586580 20.574836
C -0.654129 3.685000 20.251151
H -0.378391 4.835774 22.044905
C -1.253695 2.468068 19.752136
H -2.308911 0.676537 20.242568
H -0.217257 4.368984 19.617268
H -1.231935 2.233947 18.759012
```

21 Cartesian coordinates of monomer B (Tetracene)

```
C -3.099051 7.265949 29.291033
C -2.456426 8.147437 28.468334
C -2.400272 7.887154 27.022786
C -1.768537 8.735613 26.189052
C -1.682966 8.487143 24.780287
C -1.074191 9.323713 23.888928
C -2.936658 6.689849 26.528676
C -3.638615 5.793773 27.409001
C -3.698618 6.049017 28.792019
H -3.120811 7.500070 30.284157
H -2.043836 9.057480 28.800601
H -1.333112 9.662766 26.544615
H -0.65496  10.25623 24.251847
H -3.974355 4.898243 26.998265
H -4.135489 5.365033 29.425902
C -2.280651 7.264305 24.262882
C -1.026959 9.061599 22.514493
C -2.889426 6.427735 25.154242
C -2.195080 7.015835 22.854117
C -1.563344 7.864294 22.020383
C -0.325002 9.957675 21.634168
H -3.308652 5.495210 24.791322
H -2.630505 6.088682 22.498554
C -1.507190 7.604011 20.574836
```

```
C -0.264999 9.702431 20.251151
H 0.010739 10.853205 22.044905
C -0.864565 8.485499 19.752136
H -1.919781 6.693968 20.242568
H 0.171872 10.386415 19.617268
H -0.842805 8.251378 18.759012
’
```

22 Cartesian coordinates of monomer C (Tetracene)

```
C 0.119724 4.488661 29.164747
C 0.125554 5.377539 28.113997
C 0.767120 4.982022 26.908761
C 0.838887 5.856768 25.806516
C 1.504291 5.542182 24.585341
C 1.555408 6.389137 23.501487
C 1.354606 3.677443 26.742014
C 1.279616 2.845205 27.813607
C 0.674477 3.130571 28.990644
H -0.34293 4.825499 30.087984
H -0.30756 6.350876 28.261126
H 0.409381 6.856387 25.919315
H 1.130882 7.345483 23.638808
H 1.726696 1.867713 27.623565
H 0.634023 2.454764 29.732421
C 2.042962 4.191835 24.457829
C 2.192647 6.056573 22.301155
C 1.991845 3.344880 25.541683
C 2.708366 3.877249 23.236654
C 2.780133 4.751994 22.134408
C 2.267637 6.888812 21.229562
H 2.416371 2.388534 25.404362
H 3.137873 2.877630 23.123854
C 3.421700 4.356478 20.929173
C 2.872777 6.603446 20.052526
H 1.820558 7.866304 21.419604
C 3.427530 5.245356 19.878423
H 3.854813 3.383141 20.782043
H 2.913230 7.279253 19.310748
H 3.890193 4.908518 18.955185
```

23 Cartesian coordinates of the dimer identified in the α form of DPBF

```
O 15.670170 16.472724 26.409223
C 13.587268 13.553514 25.696234
C 14.912263 15.649764 25.620322
C 17.311043 18.217251 26.303725
C 16.137995 17.007575 24.290109
C 16.415057 17.301050 25.611952
H 13.857558 13.361218 24.827138
C 17.761806 19.383884 25.686502
C 14.041517 14.716234 26.321633
H 13.923725 15.743117 28.067999
C 13.632346 14.973933 27.634327
C 12.795139 14.089595 28.290480
H 12.534736 14.263444 29.165805
C 12.345416 12.952030 27.653987
H 11.777063 12.365080 28.097196
C 12.738551 12.686505 26.359783
```

H 12.431776 11.920116 25.930783
C 15.163086 15.947711 24.294780
H 17.465970 19.599099 24.831031
C 14.656581 15.447406 23.055273
H 14.010084 14.778283 23.042231
C 18.647000 20.222384 26.338956
H 18.950902 20.992686 25.915211
C 17.754108 17.932161 27.597345
H 17.459370 17.162418 28.027124
C 16.132054 16.985774 21.895181
H 16.451077 17.301609 21.080197
C 16.628242 17.508439 23.044567
H 17.273043 18.177562 23.022767
C 19.082135 19.923878 27.612916
H 19.680833 20.489027 28.044642
C 15.137365 15.968953 21.900825
H 14.807899 15.653677 21.089929
C 18.627375 18.786872 28.242986
H 18.909039 18.593458 29.107411
O 15.670170 22.062724 26.409223
C 14.912263 21.239764 25.620322
C 17.311043 23.807251 26.303725
C 16.137995 22.597575 24.290109
C 16.415057 22.891050 25.611952
C 15.163086 21.537711 24.294780
H 13.857558 18.951218 24.827138
C 17.761806 24.973884 25.686502
C 13.587268 19.143514 25.696234
H 12.431776 17.510116 25.930783
C 14.041517 20.306234 26.321633
C 13.632346 20.563933 27.634327
H 13.923725 21.333117 28.067999
C 12.795139 19.679595 28.290480
H 12.534736 19.853444 29.165805
C 12.345416 18.542030 27.653987
H 11.777063 17.955080 28.097196
C 12.738551 18.276505 26.359783
H 17.465970 25.189099 24.831031
C 14.656581 21.037406 23.055273
H 14.010084 20.368283 23.042231
C 18.647000 25.812384 26.338956
H 18.950902 26.582686 25.915211
C 17.754108 23.522161 27.597345
H 17.459370 22.752418 28.027124
C 16.132054 22.575774 21.895181
H 16.451077 22.891609 21.080197
C 16.628242 23.098439 23.044567
H 17.273043 23.767562 23.022767
C 19.082135 25.513878 27.612916
H 19.680833 26.079027 28.044642
C 15.137365 21.558953 21.900825
H 14.807899 21.243677 21.089929
C 18.627375 24.376872 28.242986
H 18.909039 24.183458 29.107411

24 Cartesian coordinates of the dimer identified in the β form of DPBF

O 12.456437 24.166022 26.301656
C 13.281293 23.415561 25.510705

```
C 11.931623 24.656713 24.190901
C 14.211591 22.539308 26.206858
C 11.633467 24.919862 25.510511
C 15.372287 22.088251 25.586504
H 15.566569 22.364395 24.719172
C 10.719988 25.811968 26.205695
C  9.550241 26.260296 25.589218
H  9.334426 25.964010 24.734681
C 12.989683 23.683128 24.192646
C 13.492853 23.189058 22.953101
H 14.159663 22.541127 22.933715
C 13.951890 22.122558 27.509602
H 13.180892 22.411177 27.941136
C 11.436014 25.159490 22.957172
H 10.766346 25.804171 22.941469
C 15.967051 20.835013 27.529376
H 16.550715 20.261804 27.970215
C 11.002525 26.247431 27.489828
H 11.772927 25.951015 27.917872
C 16.238686 21.235519 26.241365
H 17.006166 20.932346 25.812546
C 11.959933 24.679974 21.806609
H 11.647090 25.008878 20.995109
C  8.711286 27.143046 26.244080
H  7.938707 27.445440 25.824177
C 12.975248 23.686376 21.804283
H 13.293493 23.367609 20.991231
C  9.013352 27.573571 27.510377
H  8.450446 28.173160 27.945013
C 14.832272 21.283081 28.164075
H 14.656545 21.019413 29.038387
C 10.148912 27.118486 28.137904
H 10.342826 27.398658 29.003492
O 18.040437 24.166022 26.301656
C 18.865293 23.415561 25.510705
C 17.515623 24.656713 24.190901
C 19.795591 22.539308 26.206858
C 17.217467 24.919862 25.510511
C 20.956287 22.088251 25.586504
H 21.150569 22.364395 24.719172
C 16.303988 25.811968 26.205695
C 15.134241 26.260296 25.589218
H 14.918426 25.964010 24.734681
C 18.573683 23.683128 24.192646
C 19.076853 23.189058 22.953101
H 19.743663 22.541127 22.933715
C 19.535890 22.122558 27.509602
H 18.764892 22.411177 27.941136
C 17.020014 25.159490 22.957172
H 16.350346 25.804171 22.941469
C 21.551051 20.835013 27.529376
H 22.134715 20.261804 27.970215
C 16.586525 26.247431 27.489828
H 17.356927 25.951015 27.917872
C 21.822686 21.235519 26.241365
H 22.590166 20.932346 25.812546
C 17.543933 24.679974 21.806609
H 17.231090 25.008878 20.995109
C 14.295286 27.143046 26.244080
H 13.522707 27.445440 25.824177
```

C 18.559248 23.686376 21.804283
H 18.877493 23.367609 20.991231
C 14.597352 27.573571 27.510377
H 14.034446 28.173160 27.945013
C 20.416272 21.283081 28.164075
H 20.240545 21.019413 29.038387
C 15.732912 27.118486 28.137904
H 15.926826 27.398658 29.003492

25 Cartesian coordinates of the trimer identified in the α form of DPBF

O 18.500433 17.067276 16.676907
C 16.417530 19.986486 15.963918
H 16.687820 20.178782 15.094822
C 16.871780 18.823766 16.589317
C 16.462608 18.566067 17.902011
H 16.753987 17.796883 18.335683
C 15.625401 19.450405 18.558164
H 15.364998 19.276556 19.433489
C 15.175678 20.587970 17.921671
H 14.607326 21.174920 18.364880
C 15.568814 20.853495 16.627467
H 15.262038 21.619884 16.198467
C 17.742526 17.890236 15.888006
C 20.141305 15.322749 16.571409
C 18.968257 16.532425 14.557793
C 19.245319 16.238950 15.879636
C 17.993348 17.592289 14.562464
C 20.592069 14.156116 15.954186
H 20.296232 13.940901 15.098715
C 17.486844 18.092594 13.322957
H 16.840346 18.761717 13.309915
C 21.477262 13.317616 16.606640
H 21.781164 12.547314 16.182895
C 20.584370 15.607839 17.865029
H 20.289632 16.377582 18.294808
C 18.962316 16.554226 12.162865
H 19.281339 16.238391 11.347880
C 19.458504 16.031561 13.312251
H 20.103306 15.362438 13.290451
C 21.912397 13.616122 17.880600
H 22.511095 13.050973 18.312326
C 17.967627 17.571047 12.168509
H 17.638162 17.886323 11.357613
C 21.457638 14.753128 18.510670
H 21.739302 14.946542 19.375095
O 12.839908 17.067276 36.141539
C 10.757005 19.986486 35.428550
H 11.027295 20.178782 34.559454
C 11.211255 18.823766 36.053949
C 10.802083 18.566067 37.366643
H 11.093463 17.796883 37.800315
C 9.964876 19.450405 38.022796
H 9.704474 19.276556 38.898121
C 9.515153 20.587970 37.386303
H 8.946801 21.174920 37.829512
C 9.908289 20.853495 36.092099
H 9.601513 21.619884 35.663099
C 12.082001 17.890236 35.352638

```
C 14.480780 15.322749 36.036041
C 13.307732 16.532425 34.022425
C 13.584794 16.238950 35.344268
C 12.332823 17.592289 34.027097
C 14.931544 14.156116 35.418818
H 14.635708 13.940901 34.563347
C 11.826319 18.092594 32.787589
H 11.179822 18.761717 32.774547
C 15.816738 13.317616 36.071272
H 16.120639 12.547314 35.647527
C 14.923846 15.607839 37.329661
H 14.629107 16.377582 37.759440
C 13.301791 16.554226 31.627497
H 13.620815 16.238391 30.812513
C 13.797979 16.031561 32.776883
H 14.442781 15.362438 32.755083
C 16.251872 13.616122 37.345232
H 16.850570 13.050973 37.776958
C 12.307102 17.571047 31.633141
H 11.977637 17.886323 30.822245
C 15.797113 14.753128 37.975302
H 16.078777 14.946542 38.839727
O 15.670170 16.472724 26.409223
C 13.587268 13.553514 25.696234
H 13.857558 13.361218 24.827138
C 14.041517 14.716234 26.321633
C 13.632346 14.973933 27.634327
H 13.923725 15.743117 28.067999
C 12.795139 14.089595 28.290480
H 12.534736 14.263444 29.165805
C 12.345416 12.952030 27.653987
H 11.777063 12.365080 28.097196
C 12.738551 12.686505 26.359783
H 12.431776 11.920116 25.930783
C 14.912263 15.649764 25.620322
C 17.311043 18.217251 26.303725
C 16.137995 17.007575 24.290109
C 16.415057 17.301050 25.611952
C 15.163086 15.947711 24.294780
C 17.761806 19.383884 25.686502
H 17.465970 19.599099 24.831031
C 14.656581 15.447406 23.055273
H 14.010084 14.778283 23.042231
C 18.647000 20.222384 26.338956
H 18.950902 20.992686 25.915211
C 17.754108 17.932161 27.597345
H 17.459370 17.162418 28.027124
C 16.132054 16.985774 21.895181
H 16.451077 17.301609 21.080197
C 16.628242 17.508439 23.044567
H 17.273043 18.177562 23.022767
C 19.082135 19.923878 27.612916
H 19.680833 20.489027 28.044642
C 15.137365 15.968953 21.900825
H 14.807899 15.653677 21.089929
C 18.627375 18.786872 28.242986
H 18.909039 18.593458 29.107411
```

26 Cartesian coordinates of the trimer identified in the β form of DPBF

O 12.456437 24.166022 26.301656
C 13.281293 23.415561 25.510705
C 11.931623 24.656713 24.190901
C 14.211591 22.539308 26.206858
C 11.633467 24.919862 25.510511
C 15.372287 22.088251 25.586504
H 15.566569 22.364395 24.719172
C 10.719988 25.811968 26.205695
C 9.550241 26.260296 25.589218
H 9.334426 25.964010 24.734681
C 12.989683 23.683128 24.192646
C 13.492853 23.189058 22.953101
H 14.159663 22.541127 22.933715
C 13.951890 22.122558 27.509602
H 13.180892 22.411177 27.941136
C 11.436014 25.159490 22.957172
H 10.766346 25.804171 22.941469
C 15.967051 20.835013 27.529376
H 16.550715 20.261804 27.970215
C 11.002525 26.247431 27.489828
H 11.772927 25.951015 27.917872
C 16.238686 21.235519 26.241365
H 17.006166 20.932346 25.812546
C 11.959933 24.679974 21.806609
H 11.647090 25.008878 20.995109
C 8.711286 27.143046 26.244080
H 7.938707 27.445440 25.824177
C 12.975248 23.686376 21.804283
H 13.293493 23.367609 20.991231
C 9.013352 27.573571 27.510377
H 8.450446 28.173160 27.945013
C 14.832272 21.283081 28.164075
H 14.656545 21.019413 29.038387
C 10.148912 27.118486 28.137904
H 10.342826 27.398658 29.003492
O 13.055062 21.316478 16.608624
C 13.879918 22.066939 15.817672
C 12.530248 20.825787 14.497869
C 14.810216 22.943192 16.513826
C 12.232092 20.562638 15.817478
C 15.970912 23.394249 15.893472
H 16.165194 23.118105 15.026139
C 11.318613 19.670531 16.512663
C 10.148866 19.222204 15.896186
H 9.933051 19.518490 15.041648
C 13.588308 21.799372 14.499614
C 14.091478 22.293442 13.260069
H 14.758288 22.941373 13.240682
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H 13.779517 23.071323 18.248103
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