Supporting Information for "Optoelectronic and Excitonic Properties of Oligoacenes: Substantial Improvements from Range-Separated Time-Dependent Density Functional Theory"

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Figure SI-1. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of naphthalene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-2. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of anthracene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-3. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of tetracene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-4. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of pentacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-5. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of hexacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-6. Electron density difference maps ($\rho_{\text{excited}} - \rho_{\text{ground}}$) for the L_a and L_b excited states of heptacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent a negative density difference (depletion of density upon excitation). Both figures are plotted using the same isosurface contour value.



Figure SI-7. B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the L_a excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, Q_{mn} , give a measure of exciton delocalization between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-7 (continued). B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the L_a excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, Q_{mn} , give a measure of exciton delocalization between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-8. B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the L_b excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, Q_{mn} , give a measure of exciton delocalization between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-8 (continued). B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the L_b excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, Q_{mn} , give a measure of exciton delocalization between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-9. B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the L_a excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, P_{mn} , represent the probability amplitude of an electron-hole pair oscillation between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-9 (continued). B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the L_a excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, P_{mn} , represent the probability amplitude of an electron-hole pair oscillation between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-10. B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the L_b excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, P_{mn} , represent the probability amplitude of an electron-hole pair oscillation between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.



Figure SI-10 (continued). B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the L_b excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, P_{mn} , represent the probability amplitude of an electron-hole pair oscillation between sites *m* (*x*-axis) and *n* (*y*-axis). The color scale is given at the bottom.

Cartesian coordinates (in Å) of naphthalene (n = 2 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

2 4242914223	-0 7061098504	0 000000000
$2 \cdot 42 \cdot 42 \cdot 51 \cdot 42 \cdot 23$	0.7061000504	0.0000000000
-2.4242914225	-0.7061098504	0.0000000000
2.4242914223	0.7061098504	0.000000000
-2.4242914223	0.7061098504	0.000000000
1.2408323810	-1.3973210183	0.000000000
-1.2408323810	-1.3973210183	0.000000000
1.2408323810	1.3973210183	0.000000000
-1.2408323810	1.3973210183	0.000000000
0.000000000	-0.7144236970	0.000000000
0.000000000	0.7144236970	0.000000000
3.3653811664	-1.2403967226	0.000000000
-3.3653811664	-1.2403967226	0.000000000
3.3653811664	1.2403967226	0.000000000
-3.3653811664	1.2403967226	0.000000000
1.2397310645	-2.4806271080	0.000000000
-1.2397310645	-2.4806271080	0.000000000
1.2397310645	2.4806271080	0.000000000
-1.2397310645	2.4806271080	0.000000000
	2.4242914223 -2.4242914223 2.4242914223 -2.4242914223 1.2408323810 -1.2408323810 -1.2408323810 0.000000000 0.000000000 0.000000000 3.3653811664 -3.3653811664 -3.3653811664 1.2397310645 -1.2397310645 -1.2397310645 -1.2397310645	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$

Cartesian coordinates (in Å) of anthracene (n = 3 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

С	3.6468970622	-0.7110008858	0.000000000
С	-3.6468970622	-0.7110008858	0.000000000
С	3.6468970622	0.7110008858	0.000000000
С	-3.6468970622	0.7110008858	0.000000000
С	2.4712791339	-1.4018601205	0.000000000
C	-2.4712791339	-1.4018601205	0.0000000000
C	2.4712791339	1.4018601205	0.0000000000
C	-2.4712791339	1.4018601205	0.0000000000
C	1.2191788512	-0.7202877193	0.0000000000
Ĉ	-1.2191788512	-0.7202877193	0.0000000000
C	1.2191788512	0.7202877193	0.0000000000
C	-1.2191788512	0.7202877193	0.0000000000
C	0.000000000	-1.3985771347	0.0000000000
C	0.0000000000	1.3985771347	0.0000000000
Н	4.5898829821	-1.2418464118	0.000000000
Н	-4.5898829821	-1.2418464118	0.000000000
Н	4.5898829821	1.2418464118	0.000000000
Н	-4.5898829821	1.2418464118	0.000000000
Н	2.4708022790	-2.4850639561	0.000000000
Н	-2.4708022790	-2.4850639561	0.0000000000
Н	2.4708022790	2.4850639561	0.000000000
Н	-2.4708022790	2.4850639561	0.000000000
Η	0.000000000	-2.4826915166	0.000000000
Н	0.000000000	2.4826915166	0.000000000

Cartesian coordinates (in Å) of tetracene (n = 4 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

С	4.8707847156	-0.7134000555	0.000000000
С	-4.8707847156	-0.7134000555	0.000000000
С	4.8707847156	0.7134000555	0.000000000
С	-4.8707847156	0.7134000555	0.000000000
С	3.6987015651	-1.4041629857	0.000000000
С	-3.6987015651	-1.4041629857	0.000000000
С	3.6987015651	1.4041629857	0.000000000
С	-3.6987015651	1.4041629857	0.000000000
С	2.4413311670	-0.7237433440	0.000000000
С	-2.4413311670	-0.7237433440	0.000000000
С	2.4413311670	0.7237433440	0.000000000
С	-2.4413311670	0.7237433440	0.000000000
С	1.2311704214	-1.4013881947	0.000000000
С	-1.2311704214	-1.4013881947	0.000000000
С	1.2311704214	1.4013881947	0.000000000
С	-1.2311704214	1.4013881947	0.000000000
С	0.000000000	-0.7238765320	0.000000000
С	0.000000000	0.7238765320	0.000000000
Η	5.8146717746	-1.2425906719	0.000000000
Η	-5.8146717746	-1.2425906719	0.000000000
Η	5.8146717746	1.2425906719	0.000000000
Η	-5.8146717746	1.2425906719	0.000000000
Η	3.6986516330	-2.4873420056	0.000000000
Η	-3.6986516330	-2.4873420056	0.000000000
Η	3.6986516330	2.4873420056	0.000000000
Η	-3.6986516330	2.4873420056	0.000000000
Η	1.2315244388	-2.4853816667	0.000000000
Η	-1.2315244388	-2.4853816667	0.000000000
Η	1.2315244388	2.4853816667	0.000000000
Η	-1.2315244388	2.4853816667	0.000000000

Cartesian coordinates (in Å) of pentacene (n = 5 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

С	6.0952486157	-0.7146604735	0.0000000000
С	-6.0952486157	-0.7146604735	0.0000000000
С	6.0952486157	0.7146604735	0.0000000000
С	-6.0952486157	0.7146604735	0.0000000000
С	4.9249404729	-1.4054104710	0.0000000000
С	-4.9249404729	-1.4054104710	0.000000000
С	4.9249404729	1.4054104710	0.000000000
С	-4.9249404729	1.4054104710	0.0000000000
С	3.6649313260	-0.7256145237	0.000000000
С	-3.6649313260	-0.7256145237	0.000000000
С	3.6649313260	0.7256145237	0.0000000000
С	-3.6649313260	0.7256145237	0.0000000000
С	2.4591388656	-1.4030006551	0.000000000
С	-2.4591388656	-1.4030006551	0.000000000
С	2.4591388656	1.4030006551	0.0000000000
С	-2.4591388656	1.4030006551	0.0000000000
С	1.2219278852	-0.7263478100	0.0000000000
С	-1.2219278852	-0.7263478100	0.0000000000
С	1.2219278852	0.7263478100	0.0000000000
С	-1.2219278852	0.7263478100	0.0000000000
С	0.0000000000	-1.4034922552	0.0000000000
С	0.0000000000	1.4034922552	0.0000000000
Η	7.0395992066	-1.2429921076	0.0000000000
Η	-7.0395992066	-1.2429921076	0.0000000000
Η	7.0395992066	1.2429921076	0.0000000000
Η	-7.0395992066	1.2429921076	0.0000000000
Η	4.9251366281	-2.4885747964	0.0000000000
Η	-4.9251366281	-2.4885747964	0.0000000000
Η	4.9251366281	2.4885747964	0.0000000000
Η	-4.9251366281	2.4885747964	0.0000000000
Η	2.4597882374	-2.4869615272	0.0000000000
Η	-2.4597882374	-2.4869615272	0.0000000000
Η	2.4597882374	2.4869615272	0.0000000000
Η	-2.4597882374	2.4869615272	0.0000000000
Η	0.0000000000	-2.4873454816	0.000000000
Η	0.0000000000	2.4873454816	0.000000000

Cartesian coordinates (in Å) of hexacene (n = 6 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

С	7.3200013995	-0.7153487361	0.0000000000
Ċ	-7.3200013995	-0.7153487361	0.0000000000
C	7.3200013995	0.7153487361	0.0000000000
č	-7 3200013995	0 7153487361	0 0000000000000000000000000000000000000
C	6 1506427228	-1 4061029078	0 0000000000000000000000000000000000000
c	-6 1506427228	-1 1061029078	0.0000000000
C	6 1506427220	1 /061029078	
C	6 1506427220	1 1061020078	0.0000000000
C	4 0000010007	1.4001020070	0.0000000000
C	4.0092310037	0 7266560921	0.0000000000
C	-4.0092310037	-0.7266560821	0.0000000000
C	4.0092310037	0.7266560821	0.000000000
C	-4.8892318037	0.7266560821	0.0000000000
C	3.685/31/690	-1.4039306318	0.0000000000
C	-3.685/31/690	-1.4039306318	0.0000000000
C	3.685/31/690	1.4039306318	0.0000000000
С	-3.6857317690	1.4039306318	0.0000000000
С	2.4453431457	-0.7277631416	0.0000000000
С	-2.4453431457	-0.7277631416	0.000000000
С	2.4453431457	0.7277631416	0.000000000
С	-2.4453431457	0.7277631416	0.000000000
С	1.2280904353	-1.4047554194	0.000000000
С	-1.2280904353	-1.4047554194	0.000000000
С	1.2280904353	1.4047554194	0.000000000
С	-1.2280904353	1.4047554194	0.000000000
С	0.000000000	-0.7283252551	0.000000000
С	0.000000000	0.7283252551	0.000000000
Η	8.2646028515	-1.2432111889	0.000000000
Η	-8.2646028515	-1.2432111889	0.000000000
Η	8.2646028515	1.2432111889	0.000000000
Η	-8.2646028515	1.2432111889	0.000000000
Η	6.1509813623	-2.4892582500	0.000000000
Η	-6.1509813623	-2.4892582500	0.000000000
Η	6.1509813623	2.4892582500	0.000000000
Н	-6.1509813623	2.4892582500	0.000000000
Η	3.6865491432	-2.4878743772	0.0000000000
Η	-3.6865491432	-2.4878743772	0.0000000000
Н	3.6865491432	2.4878743772	0.0000000000
Н	-3.6865491432	2.4878743772	0.0000000000
Н	1.2283855812	-2.4885686822	0.0000000000
Н	-1.2283855812	-2.4885686822	0.0000000000
н	1.2283855812	2.4885686822	0.0000000000
Н	-1.2283855812	2.4885686822	0.0000000000

Cartesian coordinates (in Å) of heptacene (n = 7 benzene rings) optimized at the B3LYP/cc-pVTZ level of theory:

С	8.5449063012	-0.7157347725	0.000000000
С	-8.5449063012	-0.7157347725	0.000000000
С	8.5449063012	0.7157347725	0.000000000
С	-8.5449063012	0.7157347725	0.000000000
С	7.3760759541	-1.4064952292	0.000000000
С	-7.3760759541	-1.4064952292	0.000000000
С	7.3760759541	1.4064952292	0.000000000
С	-7.3760759541	1.4064952292	0.000000000
С	6.1138921927	-0.7272463444	0.000000000
С	-6.1138921927	-0.7272463444	0.000000000
С	6.1138921927	0.7272463444	0.000000000
С	-6.1138921927	0.7272463444	0.000000000
С	4.9116527936	-1.4044690962	0.000000000
С	-4.9116527936	-1.4044690962	0.000000000
С	4.9116527936	1.4044690962	0.000000000
С	-4.9116527936	1.4044690962	0.000000000
С	3.6695203070	-0.7285808809	0.000000000
С	-3.6695203070	-0.7285808809	0.000000000
С	3.6695203070	0.7285808809	0.000000000
С	-3.6695203070	0.7285808809	0.000000000
С	2.4547956302	-1.4055141475	0.000000000
С	-2.4547956302	-1.4055141475	0.000000000
С	2.4547956302	1.4055141475	0.000000000
С	-2.4547956302	1.4055141475	0.000000000
С	1.2233699067	-0.7294889613	0.000000000
С	-1.2233699067	-0.7294889613	0.000000000
С	1.2233699067	0.7294889613	0.000000000
С	-1.2233699067	0.7294889613	0.0000000000
С	0.0000000000	-1.4058348048	0.0000000000
С	0.0000000000	1.4058348048	0.0000000000
Н	9.4896469022	-1.2433334947	0.000000000
H	-9.4896469022	-1.2433334947	0.0000000000
H	9.4896469022	1.2433334947	0.0000000000
H	-9.4896469022	1.2433334947	0.0000000000
H	7.3764974714	-2.4896442426	0.0000000000
H	-/.3/649/4/14	-2.4896442426	0.0000000000
H	7.3764974714	2.4896442426	0.0000000000
H	-/.3/649/4/14	2.4896442426	0.0000000000
H	4.9125689218	-2.4884025749	0.0000000000
H	-4.9125689218	-2.4884025749	0.0000000000
H	4.9125689218	2.4884025749	0.0000000000
H	-4.9125689218	2.4884025749	0.0000000000
п u	2.400Z0/100U	-2.4093U09010	
н u	-2.40020/1880	-2.4093009010	
п u	2.40020/1000 0 /550571000	2.4073U07010 2 /002060616	
п u	-2.43323/1080	2,4093009010 _2 /806051000	
п u		-2.4090091000 2 /006051000	
п	0.00000000000000000000000000000000000	ム・40プUUJI0U0	0.00000000000