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

*Bryan M. Wong**[a] and Timothy H. Hsieh*[b]

[a] Materials Chemistry Department, Sandia National Laboratories, Livermore, California 94551

[b] Department of Physics, Massachuetts Institute of Technology, Cambridge, Massachusetts 02139

*Corresponding author. E-mail: bmwong@sandia.gov

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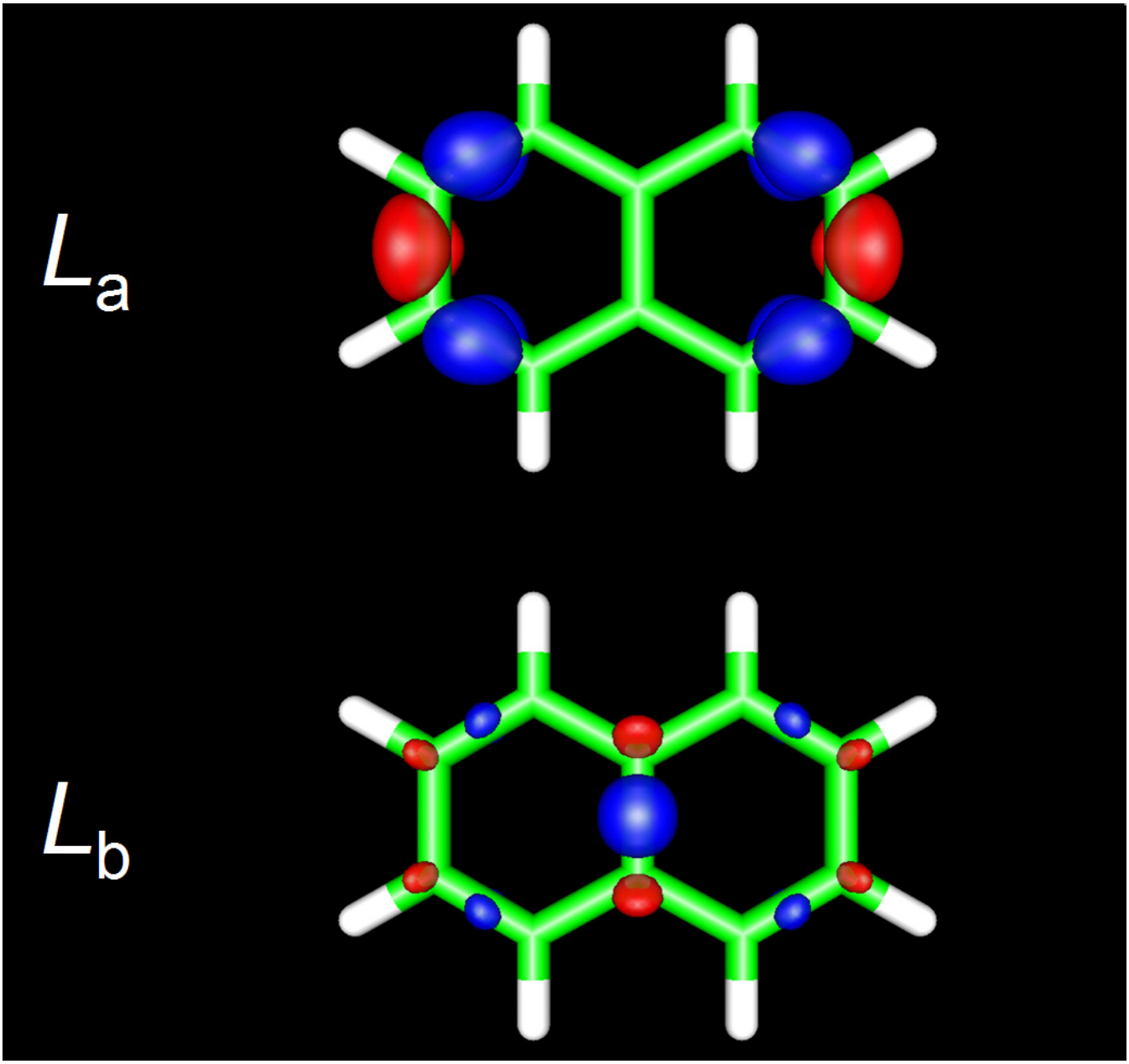


**Figure SI-1.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of naphthalene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

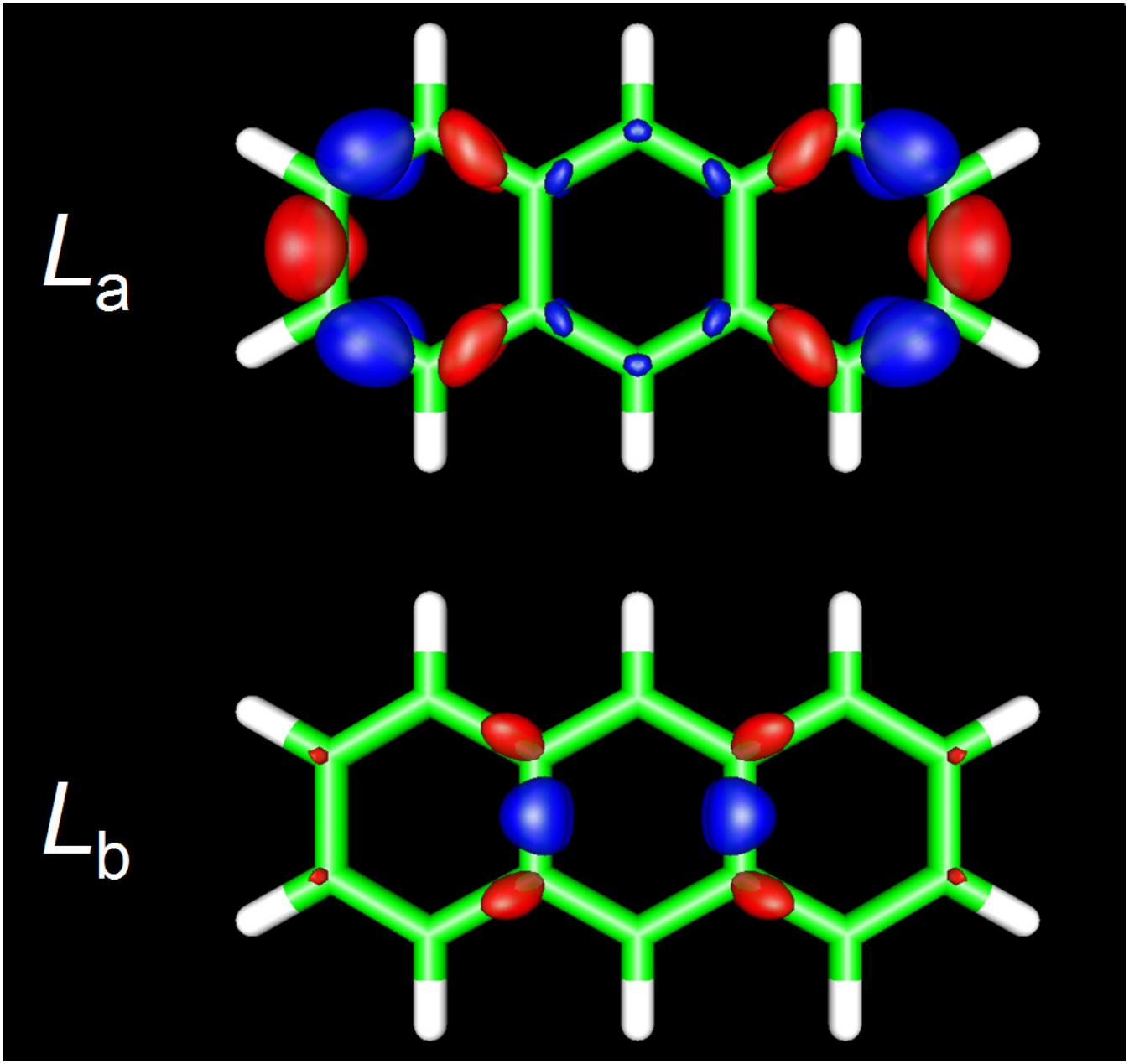


**Figure SI-2.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of anthracene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

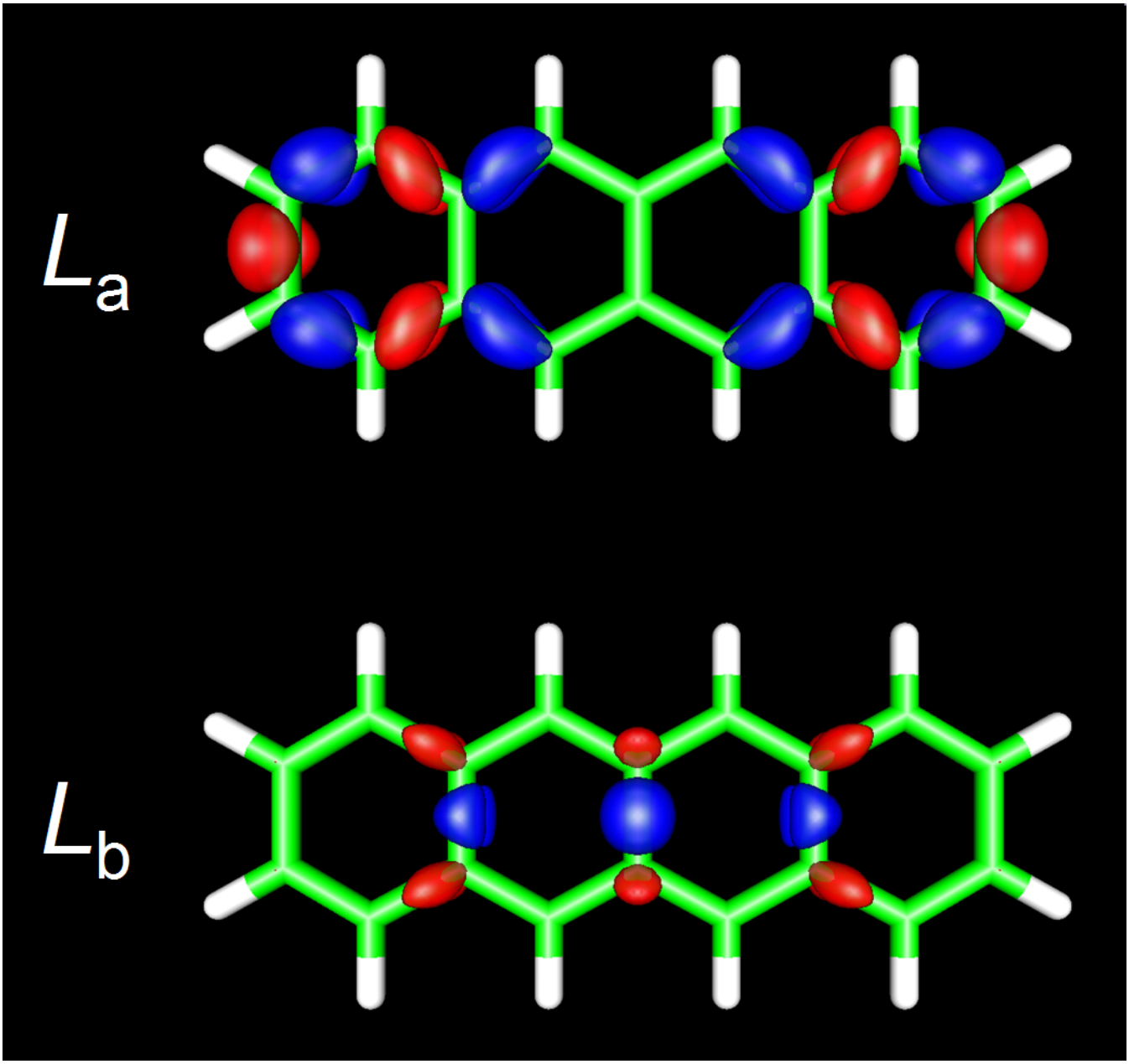


**Figure SI-3.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of tetracene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

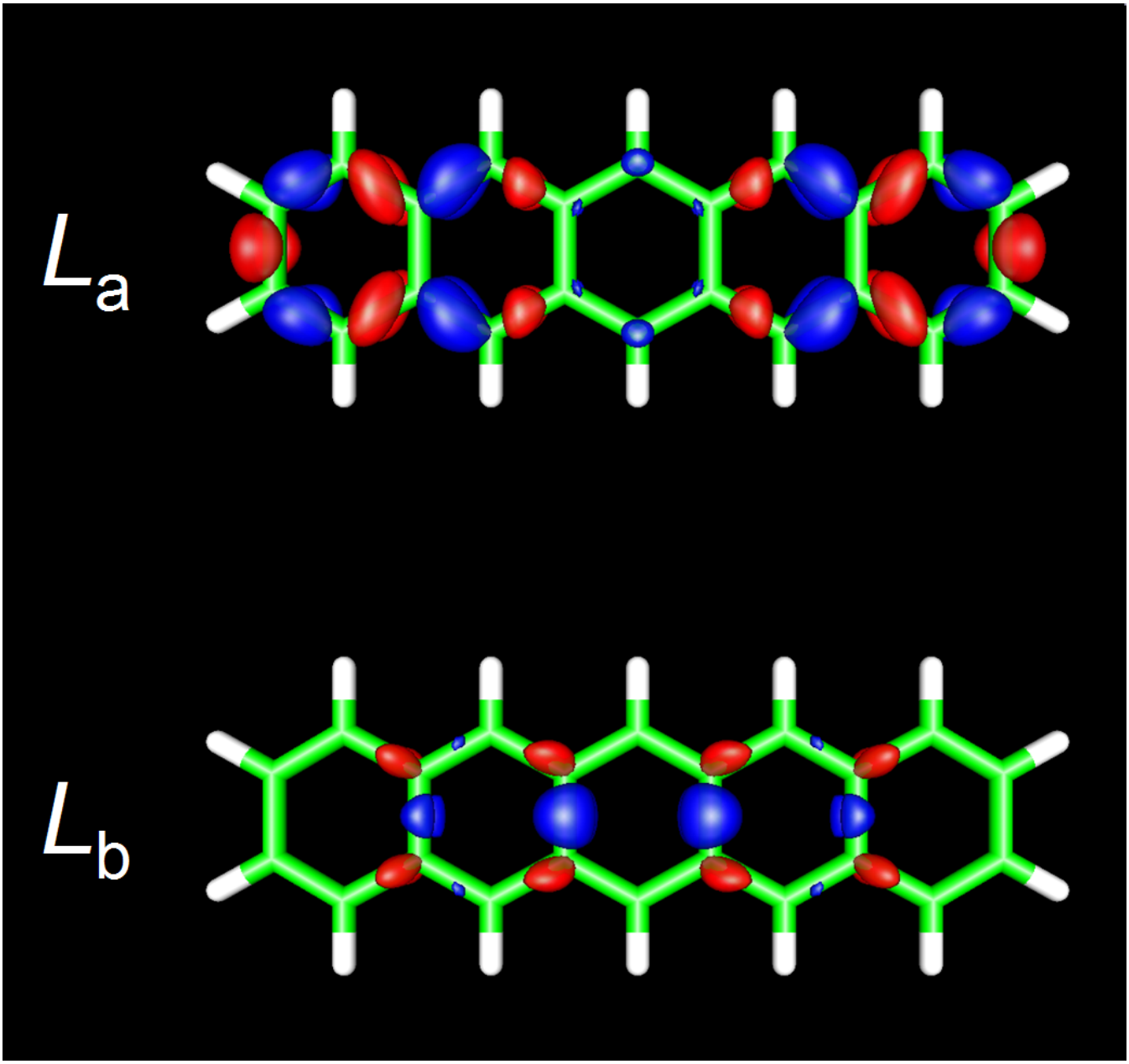


**Figure SI-4.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of pentacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

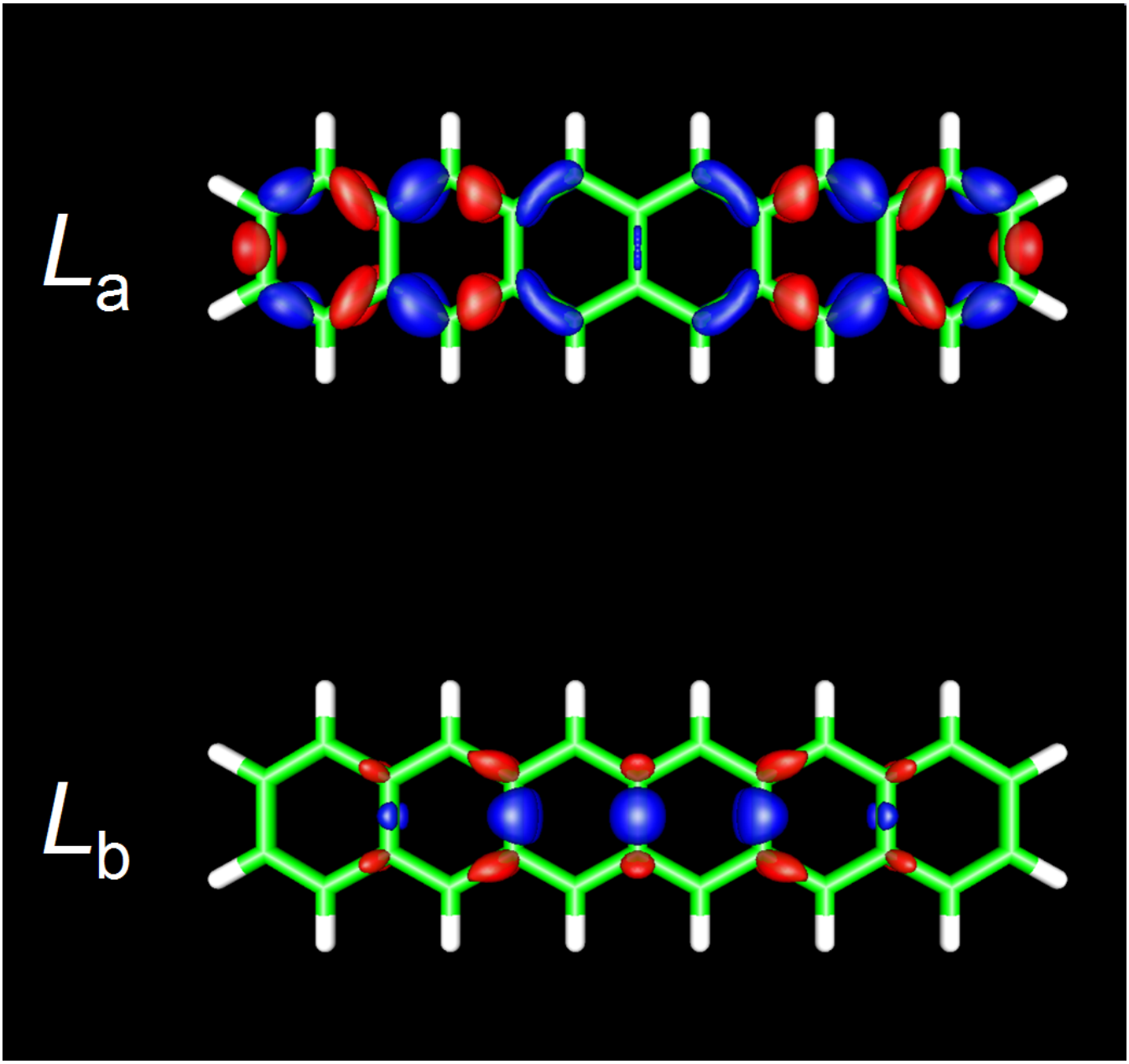


**Figure SI-5.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of hexacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

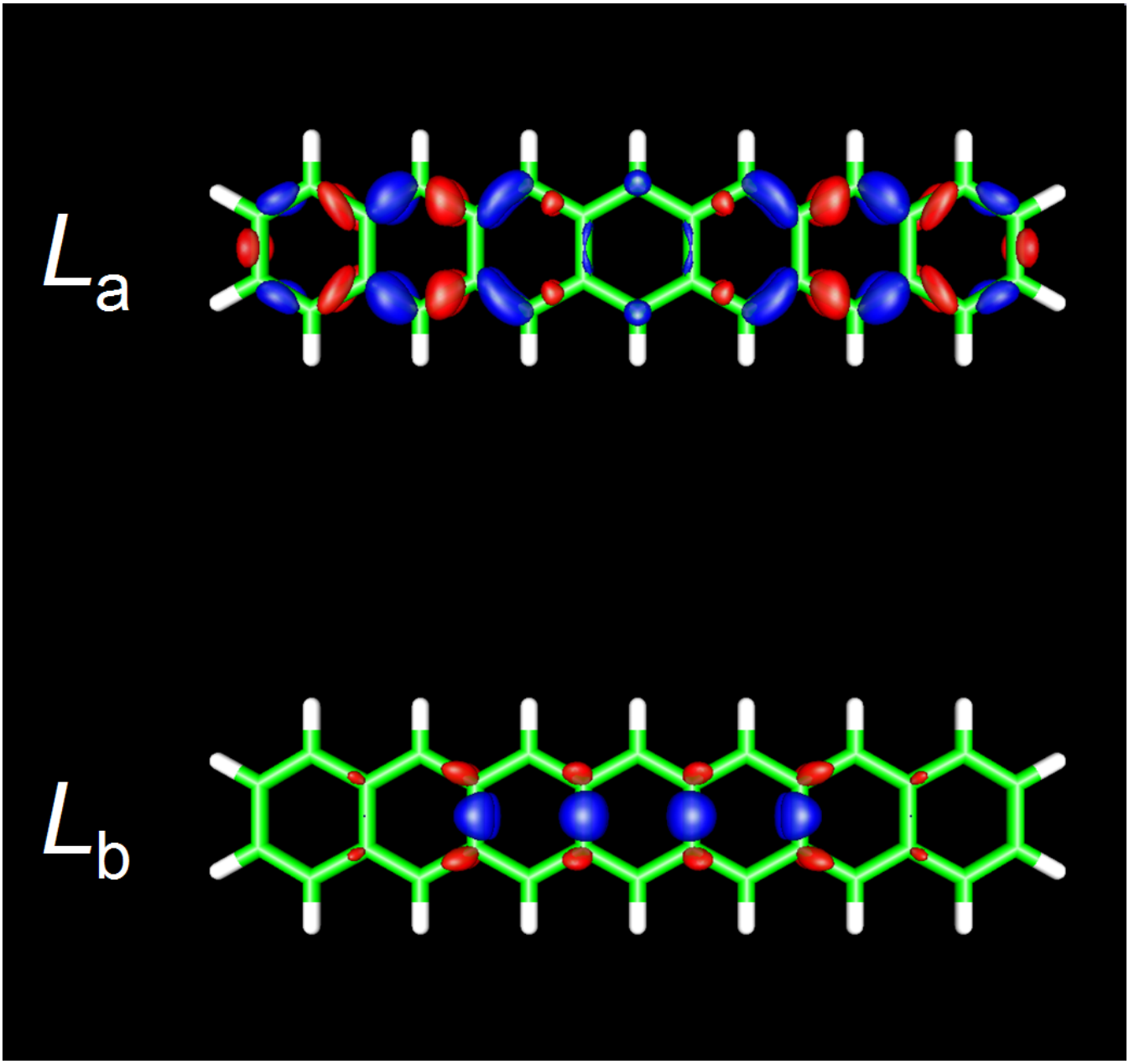


**Figure SI-6.** Electron density difference maps ($\rho_{excited} - \rho_{ground}$) for the $L_a$ and $L_b$ excited states of heptacene computed at the CC2 level of theory. Red regions denote a positive density difference (accumulation of density upon electronic excitation), and blue regions represent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

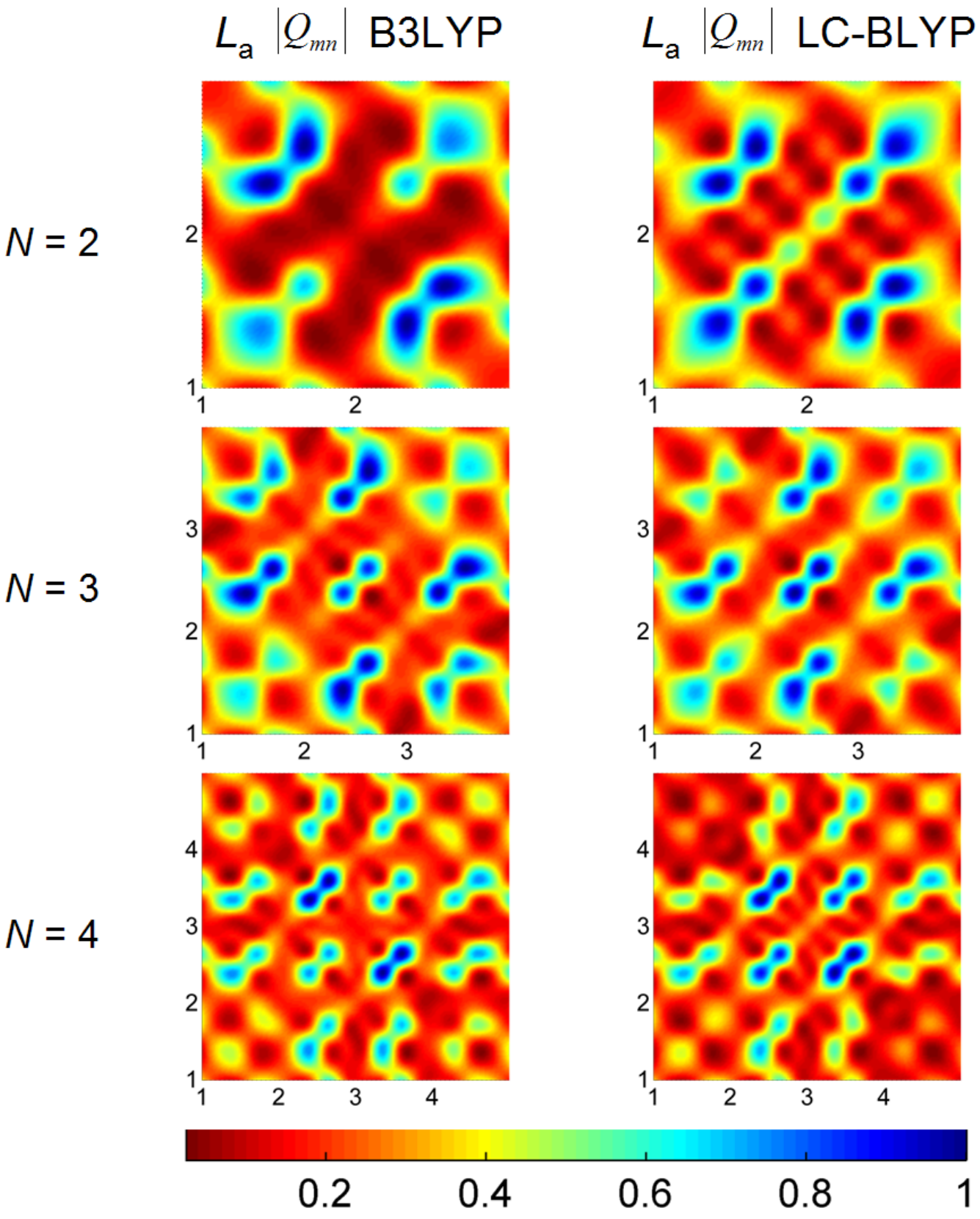


**Figure SI-7.** B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the $L_a$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, $Q_{mn}$, give a measure of exciton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

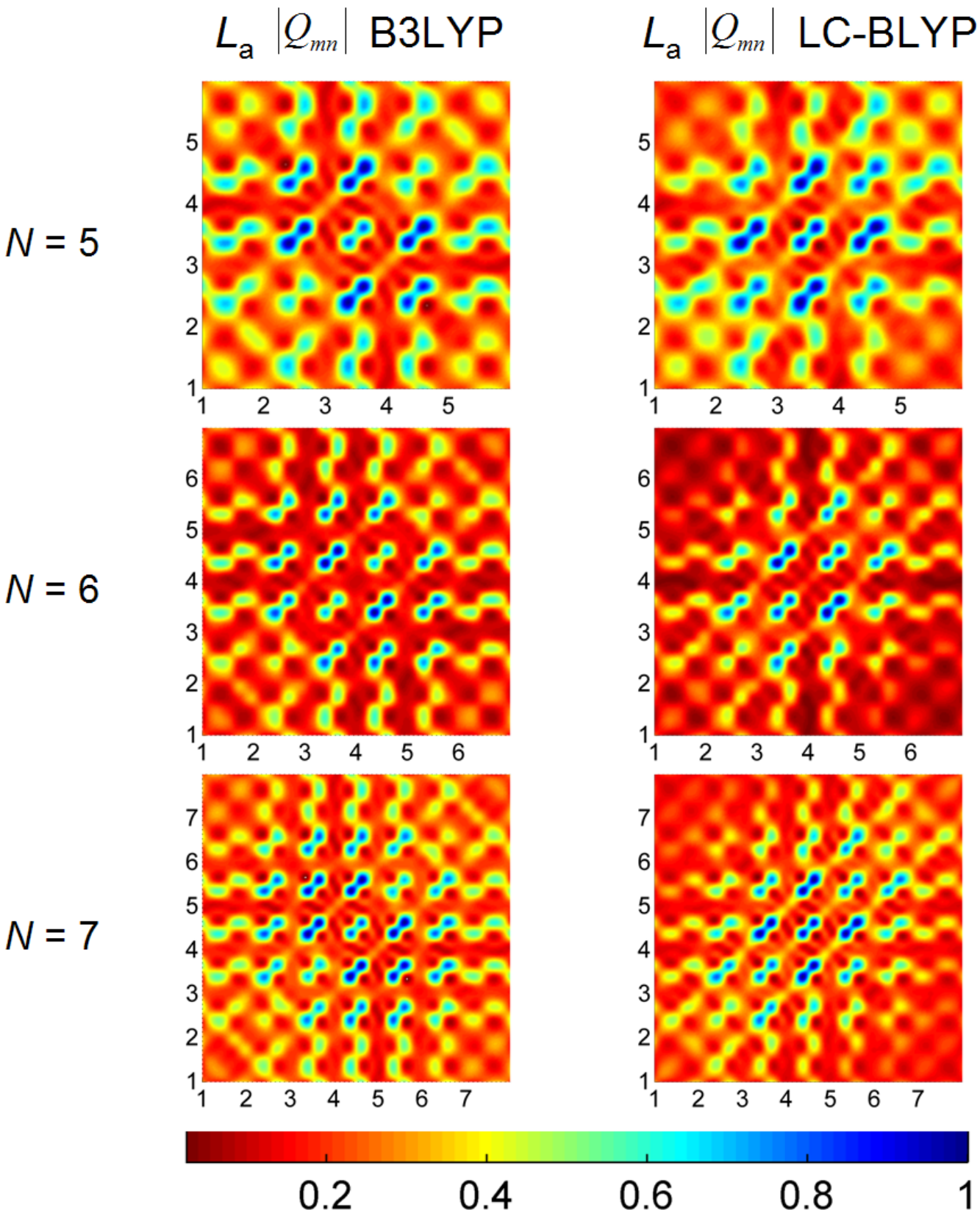


**Figure SI-7 (continued).** B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the $L_a$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, $Q_{mn}$, give a measure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

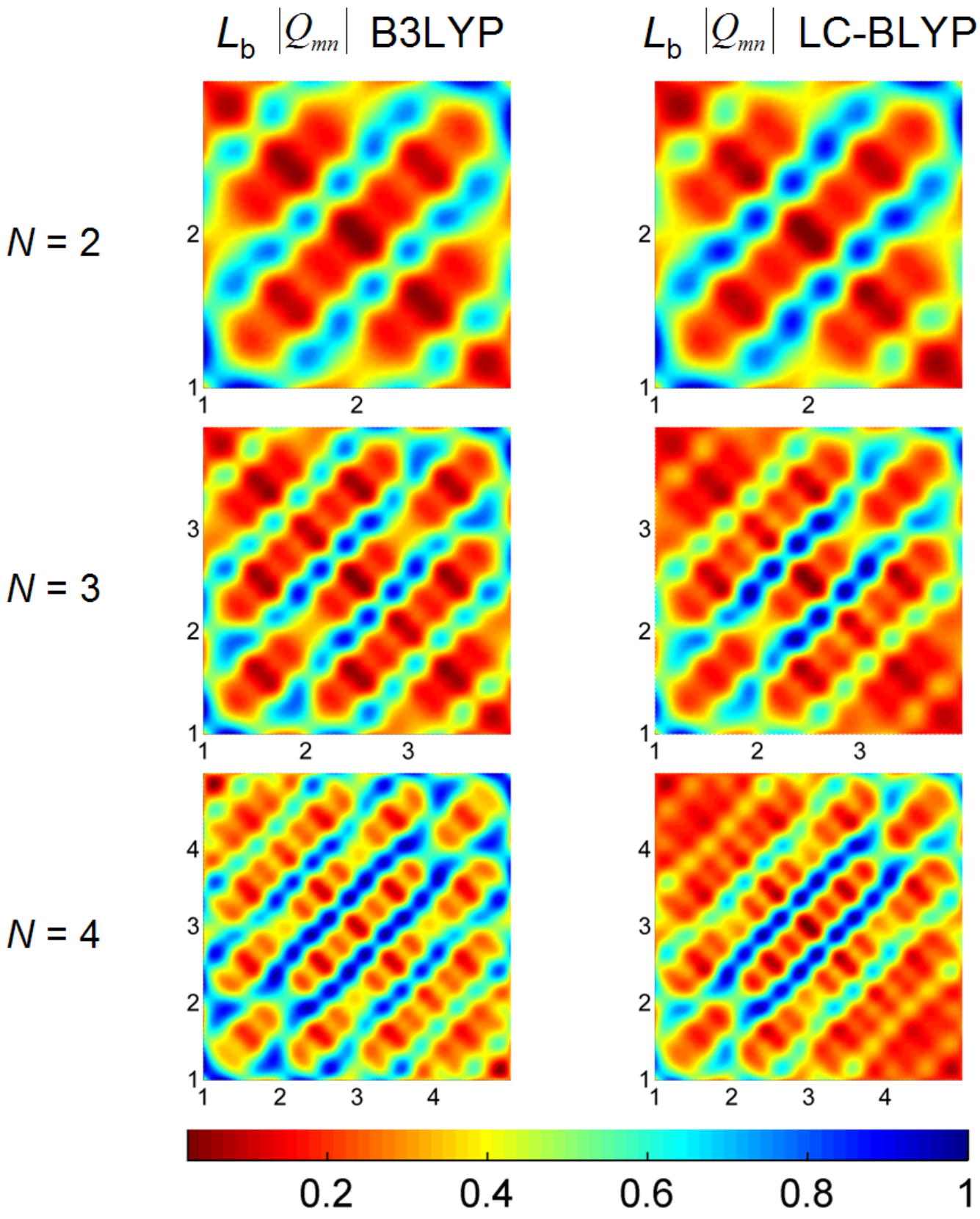


**Figure SI-8.** B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the $L_b$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, $Q_{mn}$, give a measure of exciton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

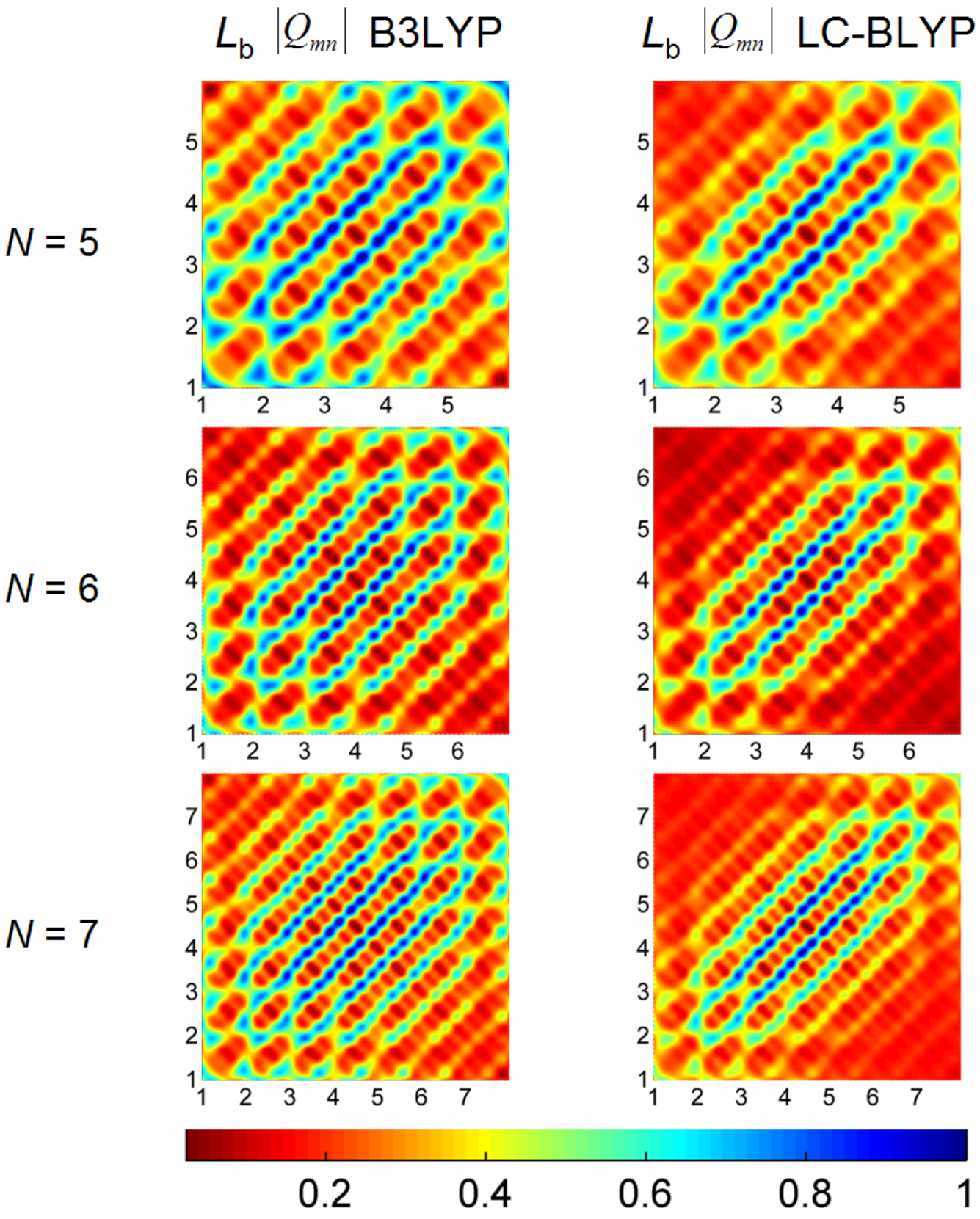


**Figure SI-8 (continued).** B3LYP and LC-BLYP contour plots of coordinate density matrices (**Q**) for the $L_b$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the coordinate matrix, $Q_{mn}$, give a measure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

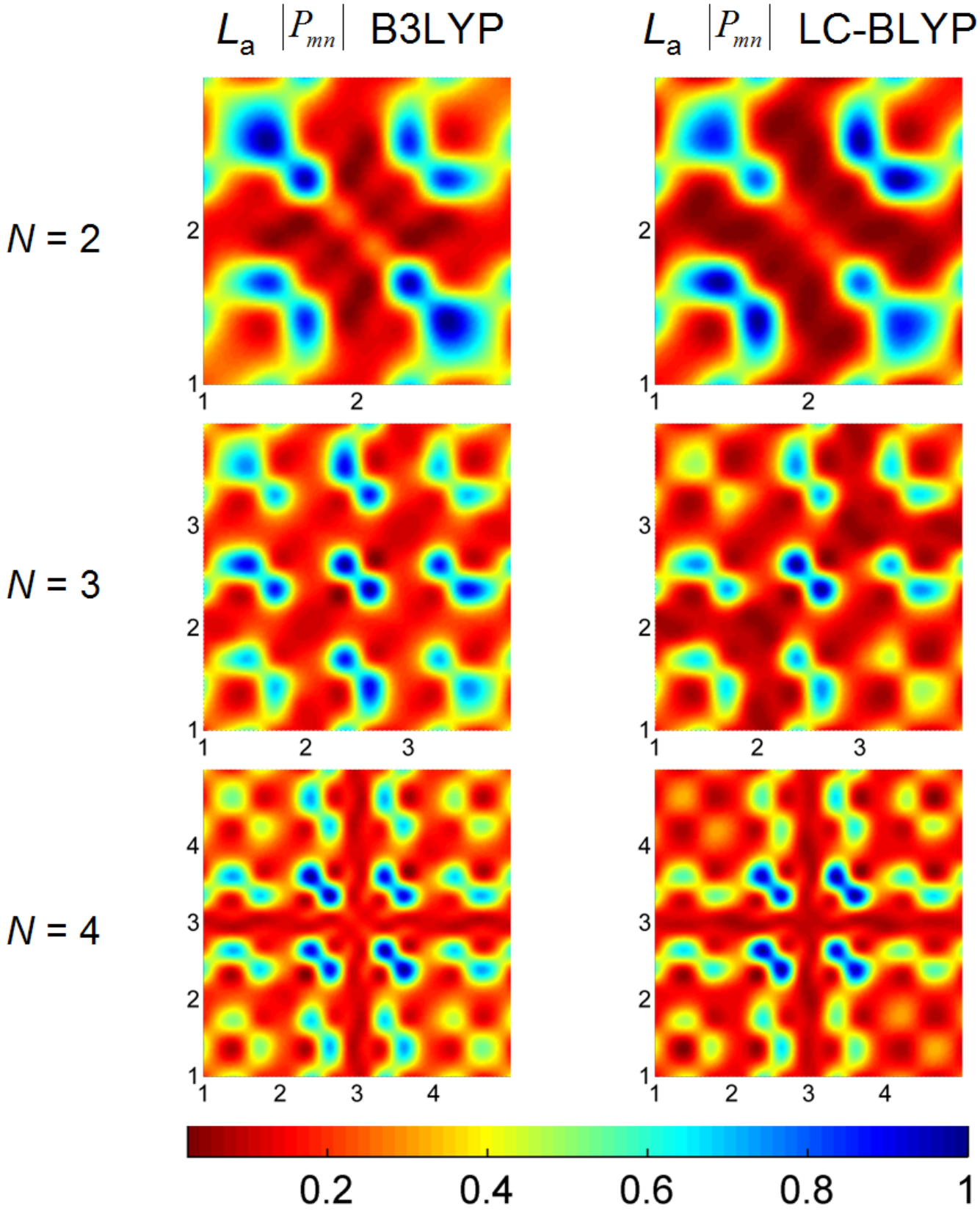


**Figure SI-9.** B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the $L_a$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, $P_{mn}$, represent the probability amplitude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

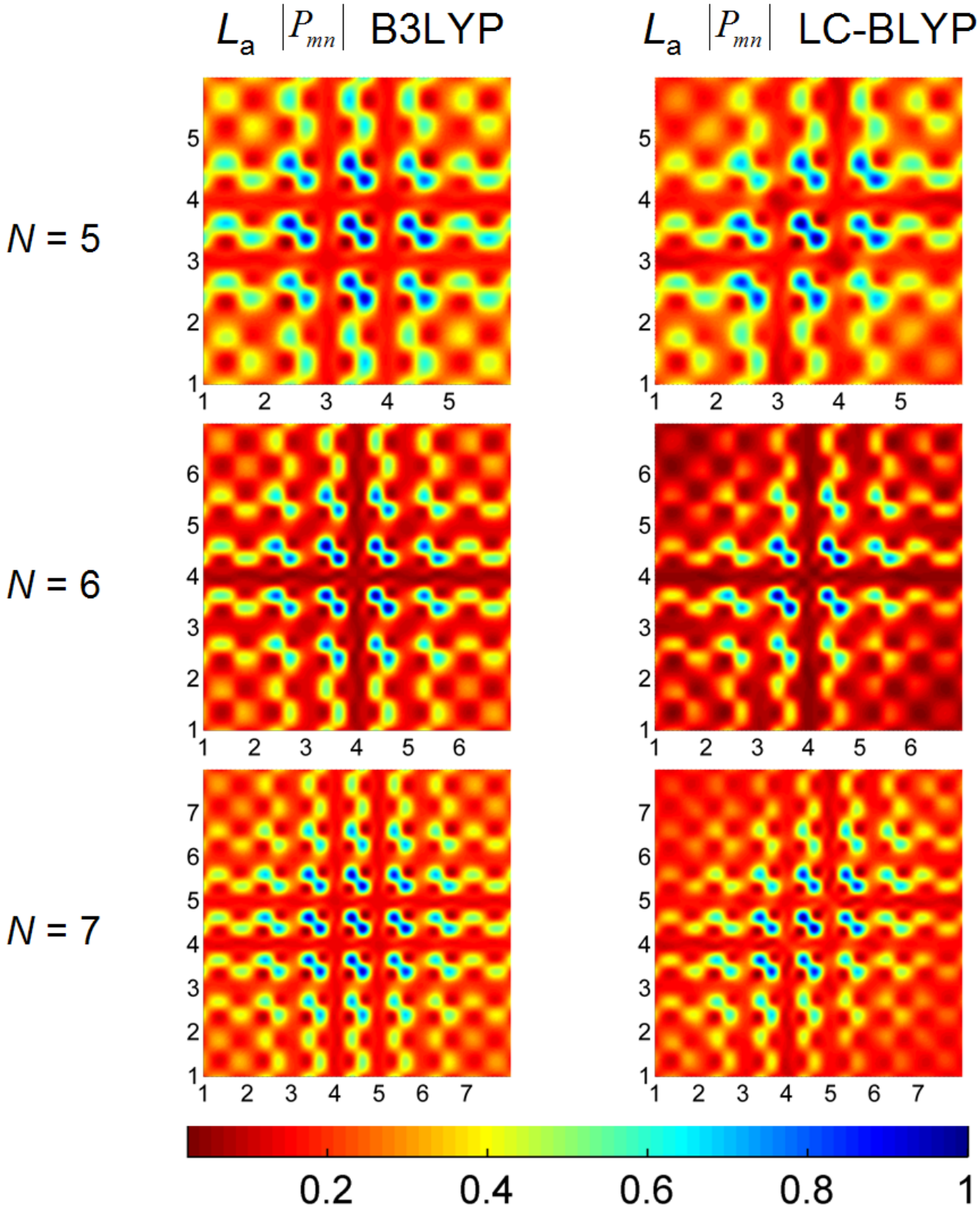


**Figure SI-9 (continued).** B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the $L_a$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, $P_{mn}$, represent the probability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

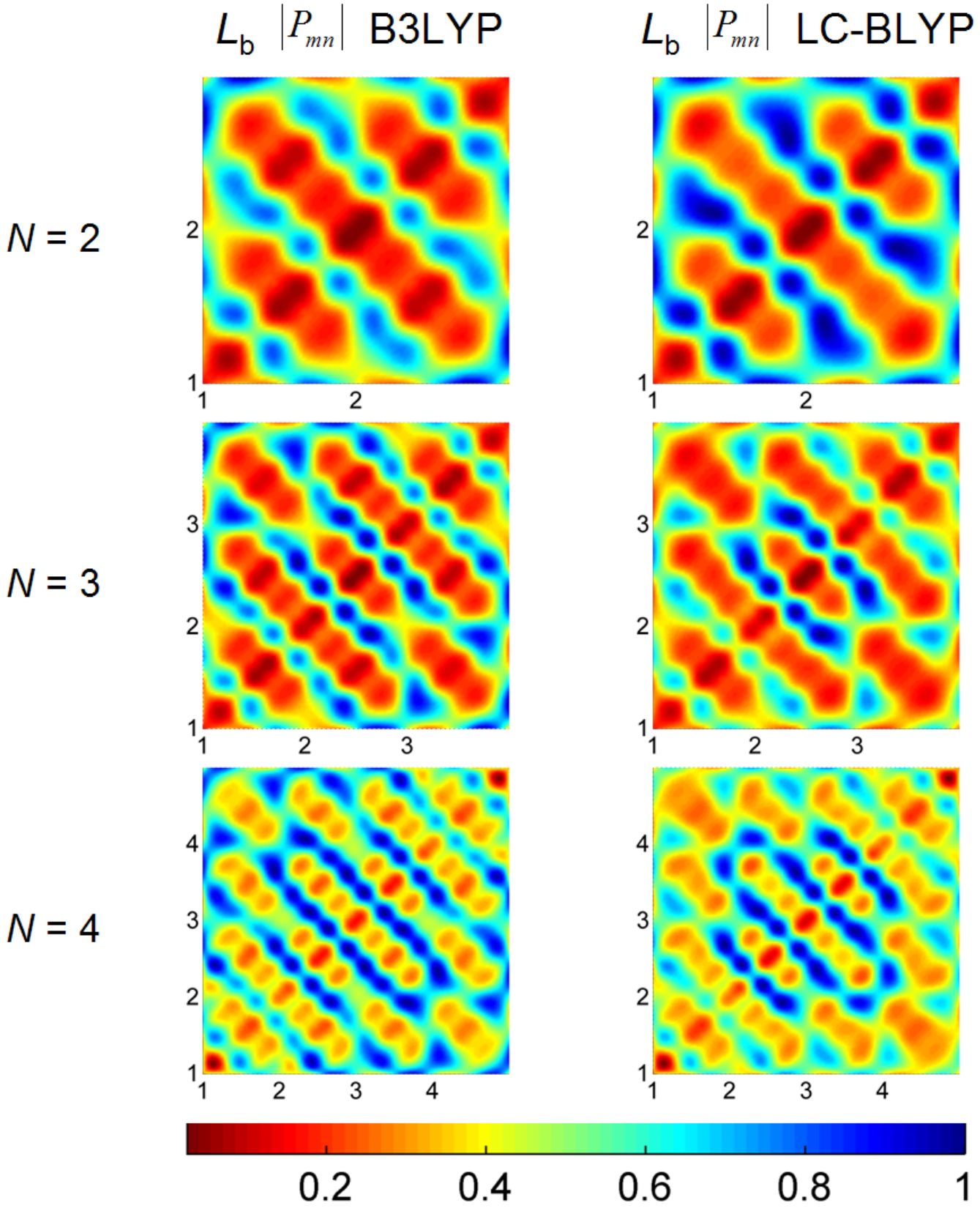


**Figure SI-10.** B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the $L_b$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, $P_{mn}$, represent the probability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

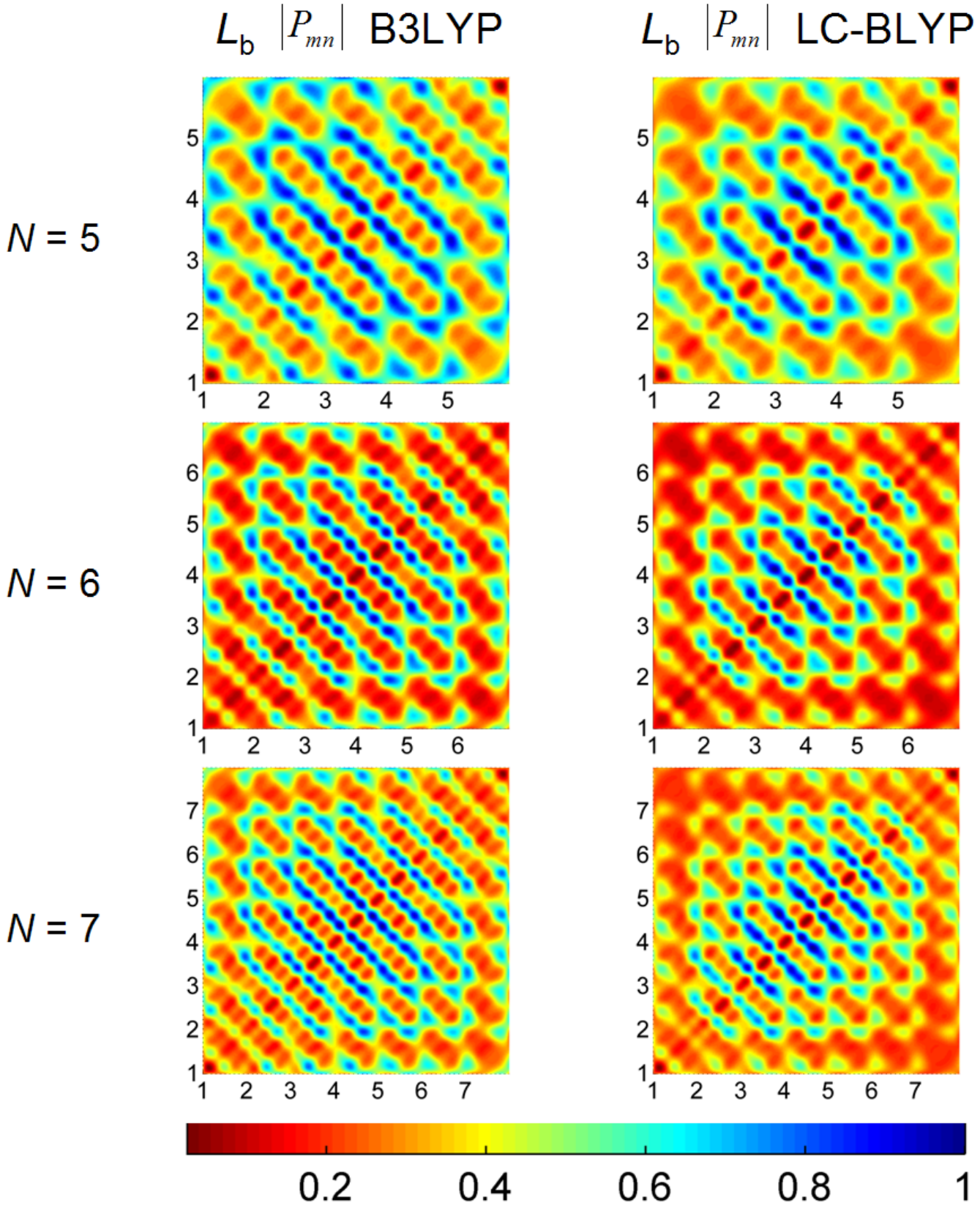


**Figure SI-10 (continued).** B3LYP and LC-BLYP contour plots of momentum density matrices (**P**) for the $L_b$ excited state in the linear acenes. The *x*- and *y*-axis labels represent the number of benzene repeat units in the molecule. The elements of the momentum matrix, $P_{mn}$, represent the 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:

| | | | |
|---|---|---|---|
| C | 2.4242914223 | -0.7061098504 | 0.0000000000 |
| C | -2.4242914223 | -0.7061098504 | 0.0000000000 |
| C | 2.4242914223 | 0.7061098504 | 0.0000000000 |
| C | -2.4242914223 | 0.7061098504 | 0.0000000000 |
| C | 1.2408323810 | -1.3973210183 | 0.0000000000 |
| C | -1.2408323810 | -1.3973210183 | 0.0000000000 |
| C | 1.2408323810 | 1.3973210183 | 0.0000000000 |
| C | -1.2408323810 | 1.3973210183 | 0.0000000000 |
| C | 0.0000000000 | -0.7144236970 | 0.0000000000 |
| C | 0.0000000000 | 0.7144236970 | 0.0000000000 |
| H | 3.3653811664 | -1.2403967226 | 0.0000000000 |
| H | -3.3653811664 | -1.2403967226 | 0.0000000000 |
| H | 3.3653811664 | 1.2403967226 | 0.0000000000 |
| H | -3.3653811664 | 1.2403967226 | 0.0000000000 |
| H | 1.2397310645 | -2.4806271080 | 0.0000000000 |
| H | -1.2397310645 | -2.4806271080 | 0.0000000000 |
| H | 1.2397310645 | 2.4806271080 | 0.0000000000 |
| H | -1.2397310645 | 2.4806271080 | 0.0000000000 |

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

| | | | |
|---|---|---|---|
| C | 3.6468970622 | -0.7110008858 | 0.0000000000 |
| C | -3.6468970622 | -0.7110008858 | 0.0000000000 |
| C | 3.6468970622 | 0.7110008858 | 0.0000000000 |
| C | -3.6468970622 | 0.7110008858 | 0.0000000000 |
| C | 2.4712791339 | -1.4018601205 | 0.0000000000 |
| 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 |
| C | -1.2191788512 | -0.7202877193 | 0.0000000000 |
| C | 1.2191788512 | 0.7202877193 | 0.0000000000 |
| C | -1.2191788512 | 0.7202877193 | 0.0000000000 |
| C | 0.0000000000 | -1.3985771347 | 0.0000000000 |
| C | 0.0000000000 | 1.3985771347 | 0.0000000000 |
| H | 4.5898829821 | -1.2418464118 | 0.0000000000 |
| H | -4.5898829821 | -1.2418464118 | 0.0000000000 |
| H | 4.5898829821 | 1.2418464118 | 0.0000000000 |
| H | -4.5898829821 | 1.2418464118 | 0.0000000000 |
| H | 2.4708022790 | -2.4850639561 | 0.0000000000 |
| H | -2.4708022790 | -2.4850639561 | 0.0000000000 |
| H | 2.4708022790 | 2.4850639561 | 0.0000000000 |
| H | -2.4708022790 | 2.4850639561 | 0.0000000000 |
| H | 0.0000000000 | -2.4826915166 | 0.0000000000 |
| H | 0.0000000000 | 2.4826915166 | 0.0000000000 |

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

| | | | |
|---|---|---|---|
| C | 4.8707847156 | -0.7134000555 | 0.0000000000 |
| C | -4.8707847156 | -0.7134000555 | 0.0000000000 |
| C | 4.8707847156 | 0.7134000555 | 0.0000000000 |
| C | -4.8707847156 | 0.7134000555 | 0.0000000000 |
| C | 3.6987015651 | -1.4041629857 | 0.0000000000 |
| C | -3.6987015651 | -1.4041629857 | 0.0000000000 |
| C | 3.6987015651 | 1.4041629857 | 0.0000000000 |
| C | -3.6987015651 | 1.4041629857 | 0.0000000000 |
| C | 2.4413311670 | -0.7237433440 | 0.0000000000 |
| C | -2.4413311670 | -0.7237433440 | 0.0000000000 |
| C | 2.4413311670 | 0.7237433440 | 0.0000000000 |
| C | -2.4413311670 | 0.7237433440 | 0.0000000000 |
| C | 1.2311704214 | -1.4013881947 | 0.0000000000 |
| C | -1.2311704214 | -1.4013881947 | 0.0000000000 |
| C | 1.2311704214 | 1.4013881947 | 0.0000000000 |
| C | -1.2311704214 | 1.4013881947 | 0.0000000000 |
| C | 0.0000000000 | -0.7238765320 | 0.0000000000 |
| C | 0.0000000000 | 0.7238765320 | 0.0000000000 |
| H | 5.8146717746 | -1.2425906719 | 0.0000000000 |
| H | -5.8146717746 | -1.2425906719 | 0.0000000000 |
| H | 5.8146717746 | 1.2425906719 | 0.0000000000 |
| H | -5.8146717746 | 1.2425906719 | 0.0000000000 |
| H | 3.6986516330 | -2.4873420056 | 0.0000000000 |
| H | -3.6986516330 | -2.4873420056 | 0.0000000000 |
| H | 3.6986516330 | 2.4873420056 | 0.0000000000 |
| H | -3.6986516330 | 2.4873420056 | 0.0000000000 |
| H | 1.2315244388 | -2.4853816667 | 0.0000000000 |
| H | -1.2315244388 | -2.4853816667 | 0.0000000000 |
| H | 1.2315244388 | 2.4853816667 | 0.0000000000 |
| H | -1.2315244388 | 2.4853816667 | 0.0000000000 |

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

| | | | |
|---|---|---|---|
| C | 6.0952486157 | -0.7146604735 | 0.0000000000 |
| C | -6.0952486157 | -0.7146604735 | 0.0000000000 |
| C | 6.0952486157 | 0.7146604735 | 0.0000000000 |
| C | -6.0952486157 | 0.7146604735 | 0.0000000000 |
| C | 4.9249404729 | -1.4054104710 | 0.0000000000 |
| C | -4.9249404729 | -1.4054104710 | 0.0000000000 |
| C | 4.9249404729 | 1.4054104710 | 0.0000000000 |
| C | -4.9249404729 | 1.4054104710 | 0.0000000000 |
| C | 3.6649313260 | -0.7256145237 | 0.0000000000 |
| C | -3.6649313260 | -0.7256145237 | 0.0000000000 |
| C | 3.6649313260 | 0.7256145237 | 0.0000000000 |
| C | -3.6649313260 | 0.7256145237 | 0.0000000000 |
| C | 2.4591388656 | -1.4030006551 | 0.0000000000 |
| C | -2.4591388656 | -1.4030006551 | 0.0000000000 |
| C | 2.4591388656 | 1.4030006551 | 0.0000000000 |
| C | -2.4591388656 | 1.4030006551 | 0.0000000000 |
| C | 1.2219278852 | -0.7263478100 | 0.0000000000 |
| C | -1.2219278852 | -0.7263478100 | 0.0000000000 |
| C | 1.2219278852 | 0.7263478100 | 0.0000000000 |
| C | -1.2219278852 | 0.7263478100 | 0.0000000000 |
| C | 0.0000000000 | -1.4034922552 | 0.0000000000 |
| C | 0.0000000000 | 1.4034922552 | 0.0000000000 |
| H | 7.0395992066 | -1.2429921076 | 0.0000000000 |
| H | -7.0395992066 | -1.2429921076 | 0.0000000000 |
| H | 7.0395992066 | 1.2429921076 | 0.0000000000 |
| H | -7.0395992066 | 1.2429921076 | 0.0000000000 |
| H | 4.9251366281 | -2.4885747964 | 0.0000000000 |
| H | -4.9251366281 | -2.4885747964 | 0.0000000000 |
| H | 4.9251366281 | 2.4885747964 | 0.0000000000 |
| H | -4.9251366281 | 2.4885747964 | 0.0000000000 |
| H | 2.4597882374 | -2.4869615272 | 0.0000000000 |
| H | -2.4597882374 | -2.4869615272 | 0.0000000000 |
| H | 2.4597882374 | 2.4869615272 | 0.0000000000 |
| H | -2.4597882374 | 2.4869615272 | 0.0000000000 |
| H | 0.0000000000 | -2.4873454816 | 0.0000000000 |
| H | 0.0000000000 | 2.4873454816 | 0.0000000000 |

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

| | | | |
|---|---|---|---|
| C | 7.3200013995 | -0.7153487361 | 0.0000000000 |
| C | -7.3200013995 | -0.7153487361 | 0.0000000000 |
| C | 7.3200013995 | 0.7153487361 | 0.0000000000 |
| C | -7.3200013995 | 0.7153487361 | 0.0000000000 |
| C | 6.1506427228 | -1.4061029078 | 0.0000000000 |
| C | -6.1506427228 | -1.4061029078 | 0.0000000000 |
| C | 6.1506427228 | 1.4061029078 | 0.0000000000 |
| C | -6.1506427228 | 1.4061029078 | 0.0000000000 |
| C | 4.8892318037 | -0.7266560821 | 0.0000000000 |
| C | -4.8892318037 | -0.7266560821 | 0.0000000000 |
| C | 4.8892318037 | 0.7266560821 | 0.0000000000 |
| C | -4.8892318037 | 0.7266560821 | 0.0000000000 |
| C | 3.6857317690 | -1.4039306318 | 0.0000000000 |
| C | -3.6857317690 | -1.4039306318 | 0.0000000000 |
| C | 3.6857317690 | 1.4039306318 | 0.0000000000 |
| C | -3.6857317690 | 1.4039306318 | 0.0000000000 |
| C | 2.4453431457 | -0.7277631416 | 0.0000000000 |
| C | -2.4453431457 | -0.7277631416 | 0.0000000000 |
| C | 2.4453431457 | 0.7277631416 | 0.0000000000 |
| C | -2.4453431457 | 0.7277631416 | 0.0000000000 |
| C | 1.2280904353 | -1.4047554194 | 0.0000000000 |
| C | -1.2280904353 | -1.4047554194 | 0.0000000000 |
| C | 1.2280904353 | 1.4047554194 | 0.0000000000 |
| C | -1.2280904353 | 1.4047554194 | 0.0000000000 |
| C | 0.0000000000 | -0.7283252551 | 0.0000000000 |
| C | 0.0000000000 | 0.7283252551 | 0.0000000000 |
| H | 8.2646028515 | -1.2432111889 | 0.0000000000 |
| H | -8.2646028515 | -1.2432111889 | 0.0000000000 |
| H | 8.2646028515 | 1.2432111889 | 0.0000000000 |
| H | -8.2646028515 | 1.2432111889 | 0.0000000000 |
| H | 6.1509813623 | -2.4892582500 | 0.0000000000 |
| H | -6.1509813623 | -2.4892582500 | 0.0000000000 |
| H | 6.1509813623 | 2.4892582500 | 0.0000000000 |
| H | -6.1509813623 | 2.4892582500 | 0.0000000000 |
| H | 3.6865491432 | -2.4878743772 | 0.0000000000 |
| H | -3.6865491432 | -2.4878743772 | 0.0000000000 |
| H | 3.6865491432 | 2.4878743772 | 0.0000000000 |
| H | -3.6865491432 | 2.4878743772 | 0.0000000000 |
| H | 1.2283855812 | -2.4885686822 | 0.0000000000 |
| H | -1.2283855812 | -2.4885686822 | 0.0000000000 |
| H | 1.2283855812 | 2.4885686822 | 0.0000000000 |
| H | -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:

| | | | |
|---|---|---|---|
| C | 8.5449063012 | -0.7157347725 | 0.0000000000 |
| C | -8.5449063012 | -0.7157347725 | 0.0000000000 |
| C | 8.5449063012 | 0.7157347725 | 0.0000000000 |
| C | -8.5449063012 | 0.7157347725 | 0.0000000000 |
| C | 7.3760759541 | -1.4064952292 | 0.0000000000 |
| C | -7.3760759541 | -1.4064952292 | 0.0000000000 |
| C | 7.3760759541 | 1.4064952292 | 0.0000000000 |
| C | -7.3760759541 | 1.4064952292 | 0.0000000000 |
| C | 6.1138921927 | -0.7272463444 | 0.0000000000 |
| C | -6.1138921927 | -0.7272463444 | 0.0000000000 |
| C | 6.1138921927 | 0.7272463444 | 0.0000000000 |
| C | -6.1138921927 | 0.7272463444 | 0.0000000000 |
| C | 4.9116527936 | -1.4044690962 | 0.0000000000 |
| C | -4.9116527936 | -1.4044690962 | 0.0000000000 |
| C | 4.9116527936 | 1.4044690962 | 0.0000000000 |
| C | -4.9116527936 | 1.4044690962 | 0.0000000000 |
| C | 3.6695203070 | -0.7285808809 | 0.0000000000 |
| C | -3.6695203070 | -0.7285808809 | 0.0000000000 |
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| C | 2.4547956302 | -1.4055141475 | 0.0000000000 |
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| C | 2.4547956302 | 1.4055141475 | 0.0000000000 |
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| C | 1.2233699067 | -0.7294889613 | 0.0000000000 |
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| C | -1.2233699067 | 0.7294889613 | 0.0000000000 |
| C | 0.0000000000 | -1.4058348048 | 0.0000000000 |
| C | 0.0000000000 | 1.4058348048 | 0.0000000000 |
| H | 9.4896469022 | -1.2433334947 | 0.0000000000 |
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| H | 9.4896469022 | 1.2433334947 | 0.0000000000 |
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| H | 7.3764974714 | -2.4896442426 | 0.0000000000 |
| H | -7.3764974714 | -2.4896442426 | 0.0000000000 |
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| H | 4.9125689218 | -2.4884025749 | 0.0000000000 |
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| H | -4.9125689218 | 2.4884025749 | 0.0000000000 |
| H | 2.4552571880 | -2.4893069616 | 0.0000000000 |
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| H | 0.0000000000 | -2.4896051808 | 0.0000000000 |
| H | 0.0000000000 | 2.4896051808 | 0.0000000000 |