

# Supporting Information for "Time-dependent open-quantum approach to two-dimensional electronic spectroscopy within a GW/BSE active space"

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# Energies and Dipole moments of Benzene

Table S1: Energies and transition dipole moments from ground state to state state of benzene. n are label of all the states computed,  $\lambda$  are the label of the states included in the time-dependent calculations of the 2D spectra.

| n  | Energy (eV) | $\mu_x$ (a.u.) | $\mu_y$ (a.u.) | $\mu_z$ (a.u.) | $\lambda$      |
|----|-------------|----------------|----------------|----------------|----------------|
| 1  | 5.1261      | 0.000000       | 0.000000       | 0.000000       |                |
| 2  | 5.1434      | 0.000000       | 0.000000       | 0.000164       |                |
| 3  | 5.3049      | 0.000000       | -0.009776      | 0.000000       |                |
| 4  | 5.6069      | 0.000000       | 0.000000       | -0.643835      | S <sub>1</sub> |
| 5  | 5.6937      | 0.000000       | 0.000000       | -0.080733      | S <sub>2</sub> |
| 6  | 5.7442      | 0.000000       | 0.000000       | 0.000000       |                |
| 7  | 5.8490      | 0.000000       | 0.000000       | 0.000000       |                |
| 8  | 5.9735      | -0.005961      | 0.000000       | 0.000000       |                |
| 9  | 6.3230      | 0.000000       | 0.000000       | 0.000000       |                |
| 10 | 6.3294      | 0.000000       | 0.000000       | 0.000129       |                |
| 11 | 6.3690      | 0.000000       | 0.000000       | 0.000060       |                |
| 12 | 6.3695      | 0.000000       | 0.000000       | 0.000000       |                |
| 13 | 6.5179      | -0.891199      | 0.000000       | 0.000000       | S <sub>3</sub> |
| 14 | 6.5411      | 0.000000       | -0.817207      | 0.000000       | S <sub>4</sub> |
| 15 | 6.7464      | 0.000000       | 0.000000       | 0.000000       |                |
| 16 | 6.7605      | 0.000000       | 0.000000       | 0.000147       |                |
| 17 | 7.2784      | -2.089893      | 0.000000       | 0.000000       | S <sub>5</sub> |
| 18 | 7.2801      | 0.000000       | 0.000000       | 0.011154       |                |
| 19 | 7.2827      | 0.000000       | 0.000265       | 0.000000       |                |
| 20 | 7.2976      | 0.000000       | 0.000000       | 0.000000       |                |
| 21 | 7.3493      | 0.000617       | 0.000000       | 0.000000       |                |
| 22 | 7.3677      | 0.000674       | 0.000000       | 0.000000       |                |

|    |        |          |           |           |                 |
|----|--------|----------|-----------|-----------|-----------------|
| 23 | 7.4128 | 0.000000 | -2.116570 | 0.000000  | S <sub>6</sub>  |
| 24 | 7.4369 | 0.000000 | 0.000000  | 0.000000  |                 |
| 25 | 7.4451 | 0.000000 | 0.000384  | 0.000000  |                 |
| 26 | 7.5351 | 0.000000 | 0.000000  | -0.154226 | S <sub>7</sub>  |
| 27 | 7.5488 | 0.000000 | 0.000000  | -0.103905 | S <sub>8</sub>  |
| 28 | 7.5697 | 0.000000 | 0.000000  | 0.000000  |                 |
| 29 | 7.7177 | 0.000000 | 0.000000  | 0.000000  |                 |
| 30 | 7.7317 | 0.000000 | 0.000000  | 0.021249  |                 |
| 31 | 7.7437 | 0.000000 | -0.000002 | 0.000000  |                 |
| 32 | 7.7504 | 0.000191 | 0.000000  | 0.000000  |                 |
| 33 | 8.0525 | 0.000000 | 0.000000  | 0.000000  |                 |
| 34 | 8.0670 | 0.000000 | 0.000000  | 0.246198  | S <sub>9</sub>  |
| 35 | 8.0677 | 0.000000 | 0.000000  | -0.014408 |                 |
| 36 | 8.1701 | 0.000000 | 0.000000  | -0.002835 |                 |
| 37 | 8.2095 | 0.000000 | 0.000000  | 0.000000  |                 |
| 38 | 8.2339 | 0.007201 | 0.000000  | 0.000000  |                 |
| 39 | 8.2580 | 0.000000 | 0.000000  | 0.000000  |                 |
| 40 | 8.3069 | 0.000000 | -0.300357 | 0.000000  | S <sub>10</sub> |
| 41 | 8.3212 | 0.356841 | 0.000000  | 0.000000  | S <sub>11</sub> |
| 42 | 8.3339 | 0.000000 | 0.000000  | -0.045448 |                 |
| 43 | 8.3607 | 0.000000 | 0.009239  | 0.000000  |                 |
| 44 | 8.3940 | 0.000000 | 0.000000  | 0.000000  |                 |
| 45 | 8.4316 | 0.000000 | 0.000000  | 0.000000  |                 |
| 46 | 8.4502 | 0.000000 | 0.000000  | 1.017812  | S <sub>12</sub> |
| 47 | 8.4947 | 0.000000 | 0.000000  | 0.000154  |                 |
| 48 | 8.5487 | 0.000000 | 0.000000  | 0.000000  |                 |
| 49 | 8.5673 | 0.000000 | 0.000000  | 0.000577  |                 |

|    |        |           |           |           |                 |
|----|--------|-----------|-----------|-----------|-----------------|
| 50 | 8.6116 | 0.000000  | 0.000000  | 0.000000  | S <sub>13</sub> |
| 51 | 8.6904 | 0.000000  | 0.000000  | -0.623679 |                 |
| 52 | 8.7795 | 0.000000  | 0.011256  | 0.000000  |                 |
| 53 | 8.8073 | -0.008661 | 0.000000  | 0.000000  |                 |
| 54 | 8.8301 | -0.000178 | 0.000000  | 0.000000  |                 |
| 55 | 8.8312 | 0.000000  | 0.000004  | 0.000000  |                 |
| 56 | 8.8544 | 0.000000  | -0.000163 | 0.000000  |                 |
| 57 | 8.8581 | -0.000254 | 0.000000  | 0.000000  |                 |
| 58 | 8.8616 | -0.049367 | 0.000000  | 0.000000  |                 |
| 59 | 8.8731 | 0.000137  | 0.000000  | 0.000000  |                 |
| 60 | 8.8965 | 0.000000  | -0.127725 | 0.000000  | S <sub>14</sub> |
| 61 | 8.9951 | 0.000000  | -0.000043 | 0.000000  |                 |
| 62 | 9.1170 | 0.000000  | 0.000000  | 0.000000  |                 |
| 63 | 9.1233 | 0.000000  | 0.000000  | -0.000050 |                 |
| 64 | 9.1265 | 0.000000  | 0.000000  | 0.000000  |                 |
| 65 | 9.1873 | 0.000000  | 0.000000  | 0.009233  |                 |
| 66 | 9.1942 | 0.000000  | 0.000000  | 0.000000  |                 |
| 67 | 9.2314 | 0.000000  | 0.000000  | -0.000062 |                 |
| 68 | 9.2569 | 0.000000  | 0.000000  | 0.000000  |                 |
| 69 | 9.2703 | 0.000000  | 0.000000  | 0.000073  |                 |
| 70 | 9.3102 | 0.000000  | -0.000530 | 0.000000  |                 |
| 71 | 9.3183 | 0.000761  | 0.000000  | 0.000000  |                 |
| 72 | 9.4499 | 0.000000  | 0.000092  | 0.000000  |                 |
| 73 | 9.4538 | -0.000124 | 0.000000  | 0.000000  |                 |
| 74 | 9.5025 | 0.000000  | 0.000000  | 0.002446  |                 |
| 75 | 9.5177 | 0.000000  | 0.000000  | 0.000000  |                 |
| 76 | 9.5444 | 0.000000  | 0.000000  | 0.000000  |                 |

|     |         |           |           |           |                 |
|-----|---------|-----------|-----------|-----------|-----------------|
| 77  | 9.5558  | 0.000000  | 0.000000  | 0.000048  |                 |
| 78  | 9.6056  | 0.000000  | 0.000000  | 0.000000  |                 |
| 79  | 9.8374  | 0.000000  | 0.000000  | -0.004183 |                 |
| 80  | 9.8390  | 0.000000  | 0.000000  | 0.000000  |                 |
| 81  | 9.8461  | 0.708814  | 0.000000  | 0.000000  | S <sub>15</sub> |
| 82  | 9.8523  | 0.000000  | -0.685685 | 0.000000  | S <sub>16</sub> |
| 83  | 9.8658  | 0.000000  | 0.000000  | 0.000000  |                 |
| 84  | 9.8993  | 0.000000  | 0.000000  | 0.000000  |                 |
| 85  | 9.9178  | 0.000000  | 0.000000  | 0.000015  |                 |
| 86  | 9.9658  | 0.000000  | 0.000135  | 0.000000  |                 |
| 87  | 9.9747  | 0.000000  | 0.000000  | 0.000000  |                 |
| 88  | 9.9846  | 0.000000  | 0.000000  | -0.000029 |                 |
| 89  | 10.0667 | 0.000000  | 0.000000  | -0.000001 |                 |
| 90  | 10.1087 | -0.426597 | 0.000000  | 0.000000  | S <sub>17</sub> |
| 91  | 10.1236 | 0.000000  | -0.430982 | 0.000000  | S <sub>18</sub> |
| 92  | 10.2469 | 0.000000  | 0.000000  | -0.735822 | S <sub>19</sub> |
| 93  | 10.2514 | 0.980873  | 0.000000  | 0.000000  | S <sub>20</sub> |
| 94  | 10.2551 | 0.000000  | -0.917355 | 0.000000  | S <sub>21</sub> |
| 95  | 10.2701 | -0.000002 | 0.121161  | 0.000000  | S <sub>22</sub> |
| 96  | 10.2711 | 0.503443  | 0.000000  | 0.000000  | S <sub>23</sub> |
| 97  | 10.2804 | 0.000000  | -0.580878 | 0.000000  | S <sub>24</sub> |
| 98  | 10.3313 | 0.000000  | 0.000000  | -0.000202 |                 |
| 99  | 10.6728 | 0.000000  | 0.000424  | 0.000000  |                 |
| 100 | 10.6827 | -0.000530 | 0.000000  | 0.000000  |                 |

# Geometry, Energies and Dipoles of chlorophyll b

Table S2: Optimized geometry of the phorphirin core of chlorophyll b.

| atom | X(Å)              | Y(Å)               | Z(Å)              |
|------|-------------------|--------------------|-------------------|
| C    | 0.10529334694944  | -16.80590882000487 | 4.83148081667821  |
| C    | 0.76060581551886  | -16.25720935655461 | 6.01137134922189  |
| N    | 2.00842155160294  | -15.85980178891825 | 5.70997972327535  |
| C    | 2.21528092975432  | -16.12133791334643 | 4.36925061205200  |
| C    | 1.00741724721432  | -16.73222132537519 | 3.81837065283380  |
| C    | 0.17219533719506  | -16.16506981852268 | 7.27766927170053  |
| C    | 0.72672937539966  | -15.64415088504209 | 8.43009385576928  |
| N    | 1.94931458272476  | -15.09812920222679 | 8.55543203936852  |
| C    | 2.12689911240555  | -14.63388966512449 | 9.84237501879451  |
| C    | 0.84429967745891  | -14.74348953639265 | 10.63718269205772 |
| C    | -0.00895288186930 | -15.66755860951192 | 9.75373290196410  |
| C    | 3.31038406226112  | -14.11758428945044 | 10.26873978630416 |
| C    | 3.70654346847429  | -13.58700176519109 | 11.64612462708964 |
| C    | 5.17986669648649  | -13.08062715150783 | 11.47879303159076 |
| C    | 5.55107376854556  | -13.35251584966810 | 10.09580014907703 |
| C    | 4.45106182902826  | -13.95434811453197 | 9.44677746178678  |
| C    | 6.55620130898598  | -13.22722555923439 | 9.16138333664618  |
| C    | 6.00180058603909  | -13.77516349264408 | 7.94935343992754  |
| N    | 4.69828062217072  | -14.21120031764822 | 8.17654078369003  |
| C    | 3.59076029985799  | -14.63793259411290 | 12.72977322824556 |
| O    | 2.67528417505556  | -14.71182417145092 | 13.50761477293534 |
| O    | 5.80765558598602  | -12.55681525150115 | 12.36565956147307 |
| Mg   | 3.34457775877403  | -15.03455152513453 | 6.95760120630170  |

|   |                   |                    |                   |
|---|-------------------|--------------------|-------------------|
| N | 4.74480078427591  | -14.89185744026671 | 5.44690391295186  |
| C | 4.55308597228726  | -15.27392115991657 | 4.17508946189924  |
| C | 5.73469153270925  | -15.00790860218553 | 3.40394132696568  |
| C | 6.66177514962491  | -14.45017339775366 | 4.26824707097466  |
| C | 6.02551353959751  | -14.38154005801301 | 5.53985628064754  |
| C | 6.59803235852171  | -13.87132647357698 | 6.70929741235899  |
| C | 7.91782338073153  | -12.65358851115808 | 9.34277190277945  |
| C | -0.11486210586204 | -17.08661452887372 | 10.31056955857346 |
| C | 0.19724010469947  | -13.37911230918209 | 10.91484168646848 |
| C | -0.03135011964498 | -12.52827981425161 | 9.67691674088548  |
| C | -0.70549799315223 | -11.22226627665925 | 9.98773133514200  |
| O | -1.06624036873658 | -10.85218423461384 | 11.07206235325590 |
| C | 3.35863445162283  | -15.86181248376470 | 3.67700424592875  |
| C | 8.03900938070620  | -13.96366991585370 | 3.94515805305967  |
| C | 8.06516476525499  | -12.50775678178727 | 3.47478648337105  |
| C | 0.86865344717502  | -17.13081697798214 | 2.42283802127930  |
| C | 0.14089710765616  | -18.14574099984388 | 1.97356187146782  |
| C | -1.30243564917180 | -17.29172996323906 | 4.79623788239204  |
| O | -0.87547457383672 | -10.47223095521488 | 8.87666225071903  |
| H | -1.31242392504015 | -9.65387043265523  | 9.16190319374512  |
| O | -0.87547457383672 | -10.47223095521488 | 8.87666225071903  |
| H | -1.31242392504015 | -9.65387043265523  | 9.16190319374512  |
| O | 4.60974080166902  | -15.50323969022205 | 12.69762581276967 |
| C | 4.57786798132766  | -16.53116876658554 | 13.69622007707032 |
| H | -0.83109866124987 | -16.55537408927524 | 7.37153236999249  |
| H | 3.38937045852093  | -16.12317175101720 | 2.63045709068080  |
| H | 7.61346154604464  | -13.50602327782047 | 6.62870732965280  |

|   |                   |                    |                   |
|---|-------------------|--------------------|-------------------|
| H | 1.03711138323177  | -15.20370441806303 | 11.60523262996338 |
| H | -1.01706569315233 | -15.26958067375070 | 9.62610735131135  |
| H | -0.66896843890678 | -17.73744162969394 | 9.63548124184372  |
| H | 0.87571178579333  | -17.51908337146733 | 10.45706708356862 |
| H | -0.62660727101602 | -17.07781445671101 | 11.27278991643811 |
| H | 0.80843411829508  | -12.82444877410560 | 11.62506373986102 |
| H | -0.75461541845278 | -13.54810820328255 | 11.41950431682040 |
| H | -0.64837063794584 | -13.03707375072304 | 8.93318593045937  |
| H | 0.90746755318623  | -12.29708439499429 | 9.16987554373368  |
| H | -1.94286029725302 | -16.68746733614810 | 5.43786401335155  |
| H | -1.69961834553759 | -17.24679700752171 | 3.78404010572583  |
| H | -1.38243943199310 | -18.32672100452611 | 5.13662619578596  |
| H | 8.48049098730079  | -14.59814133132916 | 3.17683710249087  |
| H | 8.67846565001168  | -14.07305659097559 | 4.82139167506767  |
| H | 9.08554303269329  | -12.19185391463087 | 3.25891391482367  |
| H | 7.47139684642944  | -12.37960464143922 | 2.57027151373911  |
| H | 7.65572842434884  | -11.84315323722750 | 4.23520099938989  |
| H | 8.09055708135069  | -11.82035288968925 | 8.66003367981431  |
| H | 8.04361794950890  | -12.29301544990049 | 10.36044497802670 |
| H | 8.68851056150581  | -13.40123571679654 | 9.14837704389152  |
| H | 3.09613356060823  | -12.74549048327337 | 11.97041243189306 |
| H | 1.43382579785293  | -16.54748623758357 | 1.70440535728754  |
| H | 0.09899572845792  | -18.36796620097229 | 0.91627155233005  |
| H | -0.40609435704613 | -18.80187787967853 | 2.63620848742577  |
| H | 5.47458724556126  | -17.12032183909715 | 13.53820609720104 |
| H | 4.57955541981779  | -16.08825384471540 | 14.68916224924530 |
| H | 3.68820288897115  | -17.14586599421373 | 13.57989050739187 |

|   |                  |                    |                  |
|---|------------------|--------------------|------------------|
| C | 5.94511947116458 | -15.26017076686292 | 1.98910234202376 |
| O | 5.12768065466408 | -15.72098188853005 | 1.21752615712544 |
| H | 6.95129012879958 | -14.99763017728868 | 1.61446179815412 |

Table S3: Energies and transition dipole moments from ground state to state  $\lambda$  of chlorophyll b.

| $\lambda$ | Energy (eV) | $\mu_x$ (a.u.) | $\mu_y$ (a.u.) | $\mu_z$ (a.u.) |
|-----------|-------------|----------------|----------------|----------------|
| 1         | 2.27191     | -0.83942090    | -0.67029562    | -1.26548681    |
| 2         | 2.63890     | 0.22201750     | 0.16037070     | 0.16720757     |
| 3         | 3.31365     | -2.30624632    | -0.22452618    | 2.63119143     |
| 4         | 3.47689     | -1.49786694    | -1.23753491    | -2.58434465    |
| 5         | 3.59685     | 0.41256873     | -0.14959441    | -0.87468523    |
| 6         | 3.83286     | -0.01762355    | 0.05152711     | 0.05340340     |
| 7         | 3.85531     | 0.12936982     | 0.08216821     | -0.10162678    |
| 8         | 4.00154     | -0.07376169    | 0.09628038     | 0.54051192     |
| 9         | 4.27847     | 0.92934024     | 0.28666150     | -0.35747063    |
| 10        | 4.49198     | 0.72345764     | 0.04654356     | -0.34535445    |
| 11        | 4.54959     | 0.19230037     | 0.33687086     | 0.90678891     |
| 12        | 4.63450     | 1.34913319     | 0.60582467     | 0.49736237     |
| 13        | 4.66344     | -0.30685640    | -0.39735609    | -0.94212987    |
| 14        | 4.77463     | 0.01994922     | 0.01287264     | 0.03215255     |
| 15        | 4.80940     | -0.05498257    | -0.06821796    | -0.31371502    |
| 16        | 4.95888     | -0.55347884    | -0.33291678    | -0.35164946    |
| 17        | 4.96769     | -0.11475598    | -0.07472082    | 0.05529182     |
| 18        | 4.98734     | 0.46085615     | 0.30591026     | 0.45341175     |
| 19        | 5.15603     | 0.28255555     | 0.21865706     | 0.09610092     |
| 20        | 5.16242     | -0.66517391    | -0.19409230    | 0.35175544     |

## Molecular orbitals of benzene-phenol dimer

The molecular orbitals involved in the excitations with the highest oscillator strength (states 5, 13, 15, 21) are reported in Figures S1-S4.

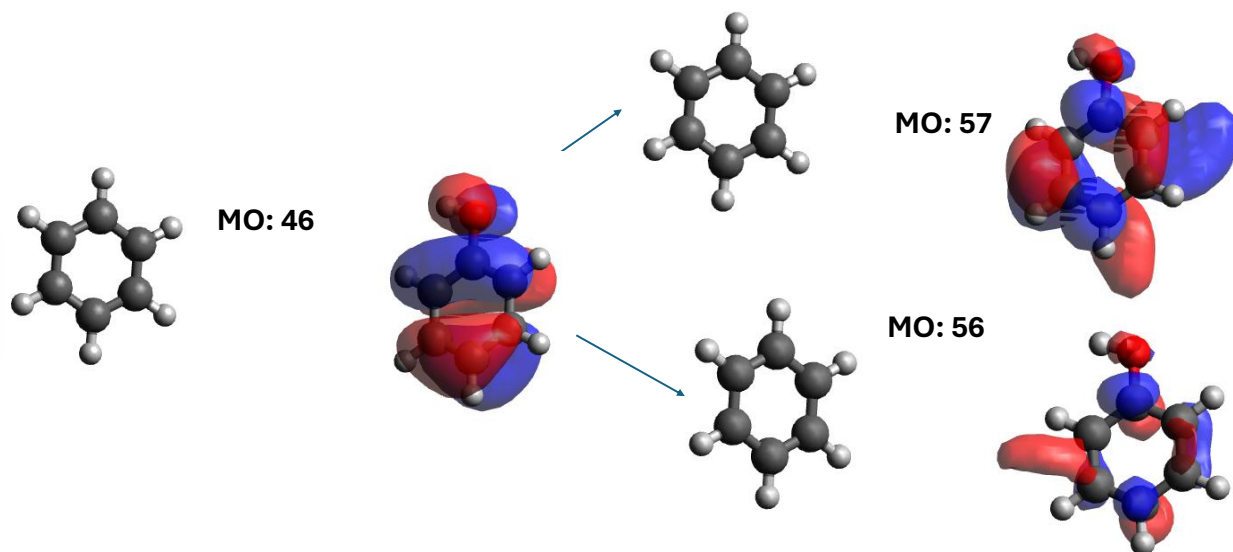


Figure S1: Molecular orbitals involved in the transitions that contribute to the 5th excited state of the benzene-phenol dimer.

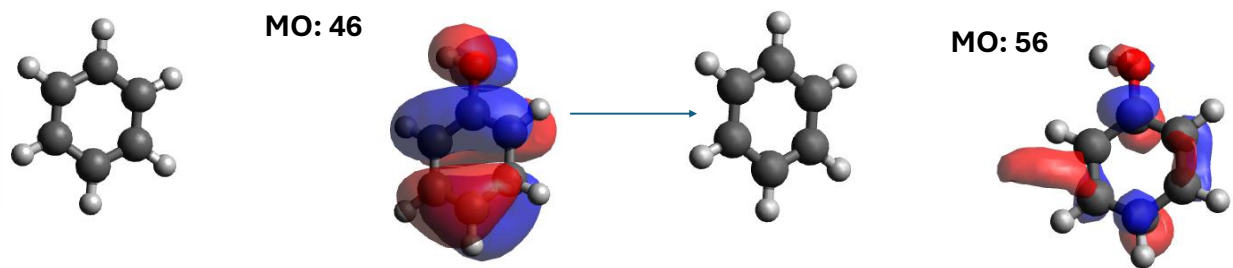


Figure S2: Molecular orbitals involved in the transitions that contribute to the 13th excited state of the benzene-phenol dimer.

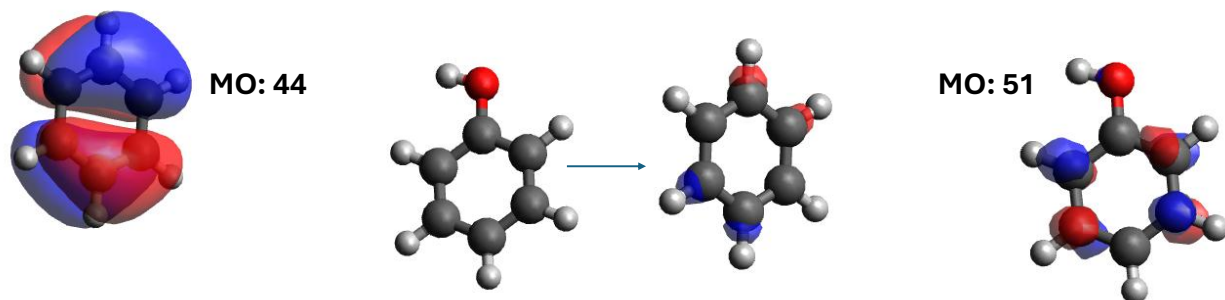


Figure S3: Molecular orbitals involved in the transitions that contribute to the 15th excited state of the benzene-phenol dimer.

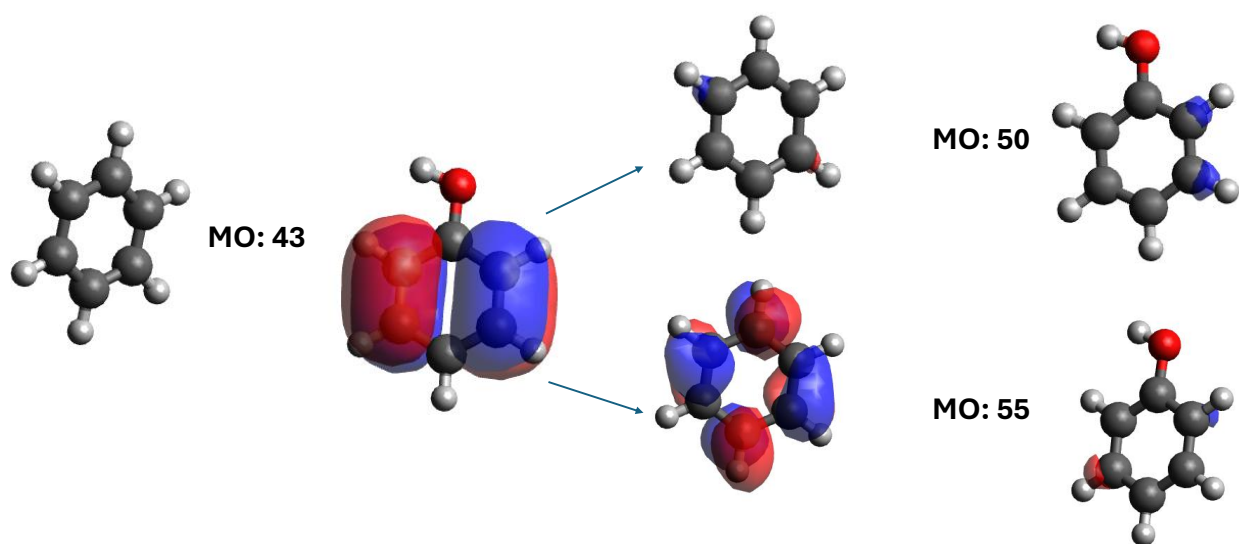


Figure S4: Molecular orbitals involved in the transitions that contribute to the 21th excited state of the benzene-phenol dimer.

# Energies and Dipole moments of benzene-phenol dimer

Table S4: Energies and transition dipole moments from ground state to state  $\lambda$  of benzene-phenol dimer.

| $\lambda$ | Energy (eV) | $\mu_x$ (a.u.) | $\mu_y$ (a.u.) | $\mu_z$ (a.u.) |
|-----------|-------------|----------------|----------------|----------------|
| 1         | 4.1248      | -0.020173      | 0.057592       | -0.010458      |
| 2         | 4.3048      | -0.091042      | 0.118039       | -0.129094      |
| 3         | 4.6797      | 0.101423       | 0.117018       | 0.085007       |
| 4         | 4.7847      | 0.158359       | 0.298608       | 0.039285       |
| 5         | 4.9744      | 0.973451       | -0.042681      | -0.320396      |
| 6         | 5.2293      | 0.060463       | 0.074250       | -0.066217      |
| 7         | 5.2301      | -0.027833      | 0.007069       | -0.017220      |
| 8         | 5.2319      | -0.022594      | 0.008571       | 0.004851       |
| 9         | 5.3102      | -0.120438      | 0.058138       | 0.086143       |
| 10        | 5.6501      | 0.072172       | -0.052364      | -0.114348      |
| 11        | 5.6922      | -0.073643      | -0.183002      | -0.160540      |
| 12        | 5.7314      | -0.501971      | 0.315821       | 0.149648       |
| 13        | 5.7630      | -0.346037      | -0.239456      | -0.058067      |
| 14        | 5.8342      | 0.432236       | -0.018384      | -0.295094      |
| 15        | 5.9358      | 0.097666       | 0.067483       | 0.585165       |
| 16        | 5.9646      | -0.060846      | 0.135893       | -0.234415      |
| 17        | 6.0273      | -0.096358      | -0.178889      | -0.113672      |
| 18        | 6.0345      | -0.076399      | 0.101541       | 0.022394       |
| 19        | 6.0728      | -0.134374      | 1.051139       | -0.742558      |
| 20        | 6.1616      | 0.032933       | -0.122774      | 0.080673       |
| 21        | 6.1665      | -0.522226      | 0.662365       | -0.940818      |
| 22        | 6.4537      | 0.339394       | 0.238713       | -0.330908      |

|    |        |           |           |           |
|----|--------|-----------|-----------|-----------|
| 23 | 6.5025 | -0.712207 | -0.200219 | 0.466660  |
| 24 | 6.5165 | 0.859776  | 0.824874  | -0.044424 |
| 25 | 6.5265 | 0.799060  | -0.775783 | -0.038268 |
| 26 | 6.5546 | -0.194999 | 0.135871  | -0.159856 |
| 27 | 6.5569 | -0.006333 | -0.011049 | 0.130326  |
| 28 | 6.5900 | 0.026256  | 0.000971  | 0.022424  |
| 29 | 6.6003 | 0.034836  | 0.077859  | -0.027113 |
| 30 | 6.6672 | -1.363894 | 0.073945  | 0.325057  |
| 31 | 6.7015 | 0.002804  | 0.039421  | 0.033374  |
| 32 | 6.7201 | -0.040015 | -0.015649 | -0.009626 |
| 33 | 6.7234 | 0.568700  | -0.009672 | -0.132512 |
| 34 | 6.7752 | -0.032791 | -0.088074 | -0.016675 |
| 35 | 6.7766 | 0.015367  | -0.076875 | -0.028403 |
| 36 | 6.7796 | -0.270338 | 0.291095  | -0.257308 |
| 37 | 6.8465 | -0.214307 | -0.091782 | 0.146136  |
| 38 | 6.8762 | -0.271318 | -0.141331 | 0.181689  |
| 39 | 6.9152 | -0.205778 | 0.237463  | -0.122681 |
| 40 | 6.9308 | 0.033123  | -0.054885 | 0.060691  |
| 41 | 6.9945 | -0.156988 | 0.013884  | 0.004867  |
| 42 | 6.9980 | -0.014565 | -0.129552 | -0.140896 |
| 43 | 7.0478 | 0.209937  | 0.294369  | -0.200148 |
| 44 | 7.1489 | 0.034667  | 0.131234  | -0.229419 |
| 45 | 7.1804 | -0.668626 | -1.724782 | 0.001103  |
| 46 | 7.2136 | 1.725306  | -0.669353 | 0.005738  |
| 47 | 7.2262 | 0.060091  | 0.000296  | -0.197922 |
| 48 | 7.2609 | -0.037628 | -0.051817 | -0.152101 |
| 49 | 7.3561 | -0.091066 | -0.179110 | 0.088856  |

|    |        |           |           |           |
|----|--------|-----------|-----------|-----------|
| 50 | 7.3642 | -0.002465 | 0.021353  | -0.062697 |
| 51 | 7.3693 | 0.080722  | -0.017609 | -0.026635 |
| 52 | 7.3970 | 0.023932  | 0.116406  | -0.005553 |
| 53 | 7.4085 | 0.013725  | -0.013428 | -0.022781 |
| 54 | 7.4193 | 0.062428  | 0.030852  | 0.009846  |
| 55 | 7.4320 | -0.150726 | -0.156177 | 0.076092  |
| 56 | 7.4414 | -0.059134 | 0.045509  | 0.099599  |
| 57 | 7.4488 | 0.150365  | -0.196015 | 0.056951  |
| 58 | 7.4731 | -0.071124 | -0.234524 | -0.102272 |
| 59 | 7.4915 | -0.001785 | 0.026164  | -0.013688 |
| 60 | 7.4990 | -0.084881 | -0.064301 | 0.160882  |
| 61 | 7.5195 | -0.021450 | 0.207459  | 0.200263  |
| 62 | 7.5382 | -0.014887 | -0.077416 | -0.134673 |
| 63 | 7.5585 | -0.012098 | 0.031165  | 0.015809  |
| 64 | 7.5658 | 0.101307  | -0.184200 | 0.248634  |
| 65 | 7.5920 | 0.059045  | -0.176985 | 0.060886  |
| 66 | 7.6421 | 0.009642  | -0.020402 | 0.015931  |
| 67 | 7.6473 | -0.028953 | -0.017008 | -0.117899 |
| 68 | 7.6530 | 0.125882  | -0.065327 | 0.012091  |
| 69 | 7.7061 | -0.060849 | -0.045950 | -0.154025 |
| 70 | 7.7562 | 0.056313  | -0.098000 | 0.088299  |
| 71 | 7.7609 | -0.053178 | -0.071324 | 0.032606  |
| 72 | 7.7673 | 0.008174  | 0.116488  | 0.120636  |
| 73 | 7.7765 | -0.104607 | -0.059326 | -0.025406 |
| 74 | 7.8053 | -0.077353 | -0.106198 | 0.014022  |
| 75 | 7.8316 | 0.076396  | 0.073078  | -0.018474 |
| 76 | 7.8503 | 0.006133  | 0.033780  | 0.003943  |

|     |        |           |           |           |
|-----|--------|-----------|-----------|-----------|
| 77  | 7.8579 | -0.002922 | -0.008388 | 0.000101  |
| 78  | 7.8761 | -0.039639 | -0.043936 | -0.037938 |
| 79  | 7.9018 | -0.003216 | -0.001069 | -0.006788 |
| 80  | 7.9028 | -0.013595 | 0.000289  | -0.000813 |
| 81  | 7.9122 | 0.044918  | -0.053828 | -0.008004 |
| 82  | 7.9596 | 0.045612  | -0.034099 | 0.086014  |
| 83  | 7.9781 | -0.009300 | 0.096430  | -0.194413 |
| 84  | 7.9886 | 0.233869  | -0.251016 | 0.086053  |
| 85  | 8.0130 | -0.073642 | -0.119585 | -0.137554 |
| 86  | 8.0288 | 0.006042  | -0.081416 | 0.109983  |
| 87  | 8.0413 | 0.061595  | 0.019670  | 0.034761  |
| 88  | 8.0599 | -0.061909 | 0.020766  | 0.008868  |
| 89  | 8.0813 | -0.007670 | -0.108469 | 0.030577  |
| 90  | 8.0844 | -0.116718 | 0.063486  | -0.056288 |
| 91  | 8.1063 | 0.000202  | 0.026494  | -0.176968 |
| 92  | 8.1389 | 0.058368  | 0.020458  | 0.003390  |
| 93  | 8.1598 | 0.122743  | -0.526151 | 0.631186  |
| 94  | 8.2135 | -0.191263 | -0.246182 | -0.064850 |
| 95  | 8.2275 | 0.038269  | -0.178824 | 0.047440  |
| 96  | 8.2453 | -0.119098 | 0.141437  | 0.107523  |
| 97  | 8.2564 | -0.021506 | -0.027685 | -0.077571 |
| 98  | 8.2592 | -0.651131 | 0.043682  | 0.274004  |
| 99  | 8.3035 | 0.048962  | -0.026289 | 0.072533  |
| 100 | 8.3201 | -0.022976 | 0.006338  | 0.011427  |
| 101 | 8.3231 | -0.018363 | 0.187947  | -0.137152 |
| 102 | 8.3636 | -0.399426 | -0.026400 | 0.060226  |
| 103 | 8.4009 | 0.028145  | 0.057284  | 0.064370  |

|     |        |           |           |           |
|-----|--------|-----------|-----------|-----------|
| 104 | 8.4114 | 0.008962  | -0.010573 | 0.004588  |
| 105 | 8.4140 | -0.002414 | -0.010607 | 0.204446  |
| 106 | 8.4252 | -0.004198 | 0.008314  | 0.021040  |
| 107 | 8.4300 | 0.003090  | -0.050155 | -0.019686 |
| 108 | 8.4353 | -0.011062 | 0.023895  | -0.133693 |
| 109 | 8.4445 | -0.014063 | 0.006124  | -0.029362 |
| 110 | 8.4539 | 0.345781  | 0.022897  | 0.013981  |
| 111 | 8.4696 | 0.156614  | 0.392085  | 0.195287  |
| 112 | 8.4722 | 0.041370  | 0.084161  | 0.698337  |
| 113 | 8.4933 | -0.029521 | -0.001610 | 0.017164  |
| 114 | 8.5128 | -0.104364 | -0.245678 | -0.071486 |
| 115 | 8.5216 | 0.051961  | 0.005066  | -0.188263 |
| 116 | 8.5399 | -0.043296 | 0.051314  | -0.686543 |
| 117 | 8.5532 | -0.189757 | 0.024564  | 0.102414  |
| 118 | 8.6133 | 0.054925  | -0.181027 | -0.251112 |
| 119 | 8.6171 | -0.039437 | -0.330815 | -0.265838 |
| 120 | 8.6207 | -0.137106 | -0.087119 | -0.016803 |
| 121 | 8.6259 | -0.003434 | 0.088962  | 0.130068  |
| 122 | 8.6397 | 0.056403  | -0.363944 | 0.084823  |
| 123 | 8.6515 | 0.246800  | 0.179965  | -0.124924 |
| 124 | 8.6518 | -0.342292 | 0.160253  | 0.017799  |
| 125 | 8.6639 | -0.147997 | 0.083784  | -0.112250 |
| 126 | 8.6646 | -0.013797 | -0.043650 | 0.014298  |
| 127 | 8.6938 | -0.002757 | 0.047846  | 0.101698  |
| 128 | 8.6992 | -0.006485 | -0.015938 | -0.019301 |
| 129 | 8.7169 | -0.028070 | -0.018962 | -0.066408 |
| 130 | 8.7301 | 0.072053  | 0.026781  | 0.168282  |

|     |        |           |           |           |
|-----|--------|-----------|-----------|-----------|
| 131 | 8.7331 | -0.032039 | -0.192542 | -0.127043 |
| 132 | 8.7493 | 0.027713  | -0.015075 | -0.373649 |
| 133 | 8.7505 | -0.009527 | -0.029645 | 0.067844  |
| 134 | 8.7605 | -0.004017 | -0.073914 | -0.271462 |
| 135 | 8.7643 | 0.003641  | 0.094784  | -0.268333 |
| 136 | 8.8028 | 0.014759  | -0.017619 | 0.030368  |
| 137 | 8.8099 | 0.056289  | -0.010844 | -0.128841 |
| 138 | 8.8117 | 0.040464  | 0.057389  | 0.003026  |
| 139 | 8.8442 | -0.050378 | 0.030196  | -0.038199 |
| 140 | 8.8683 | 0.031155  | -0.021015 | -0.012042 |
| 141 | 8.8910 | -0.003035 | 0.005719  | 0.013264  |
| 142 | 8.9035 | 0.040786  | 0.071008  | -0.018607 |
| 143 | 8.9115 | 0.006275  | -0.002268 | -0.024830 |
| 144 | 8.9140 | -0.005947 | -0.018307 | -0.030972 |
| 145 | 8.9616 | -0.031050 | 0.013812  | -0.009714 |
| 146 | 8.9697 | -0.001574 | -0.013942 | 0.031894  |
| 147 | 8.9736 | -0.009814 | -0.000844 | 0.001215  |
| 148 | 8.9859 | -0.182682 | -0.183960 | 0.037521  |
| 149 | 9.0018 | -0.018812 | -0.011157 | 0.066602  |
| 150 | 9.0214 | 0.006104  | -0.081897 | -0.253181 |

## Convergence of the number of SSE trajectories

We compared the time evolution of two benzene–phenol dimer coherences, corresponding to the pairs of states (12–15) and (13–15), to assess whether the number of trajectories considered is sufficient to accurately describe the system dynamics. These simulations were performed including the five excited states of the benzene–phenol system used in the SSE dynamics reported in the main text (Section 4.3.1). The system was excited by a single laser pulse with the same temporal profile as that described in the main text (intensity of  $I = 5.01 \times 10^9 \text{ W/cm}^2$ , temporal width of  $\delta = 2 \text{ fs}$  and a central frequency of  $\omega = 5.47 \text{ eV}$ ).

The results obtained by averaging over 100 and 200 trajectories are shown in Figure S5. In particular, the comparison between panels (b) and (d) shows that the time evolution of the coherences until 100 fs after interaction with the incident pulse is nearly identical in both cases, confirming that the use of 100 independent SSE trajectories provides a reliable statistical sampling of the system dynamics.

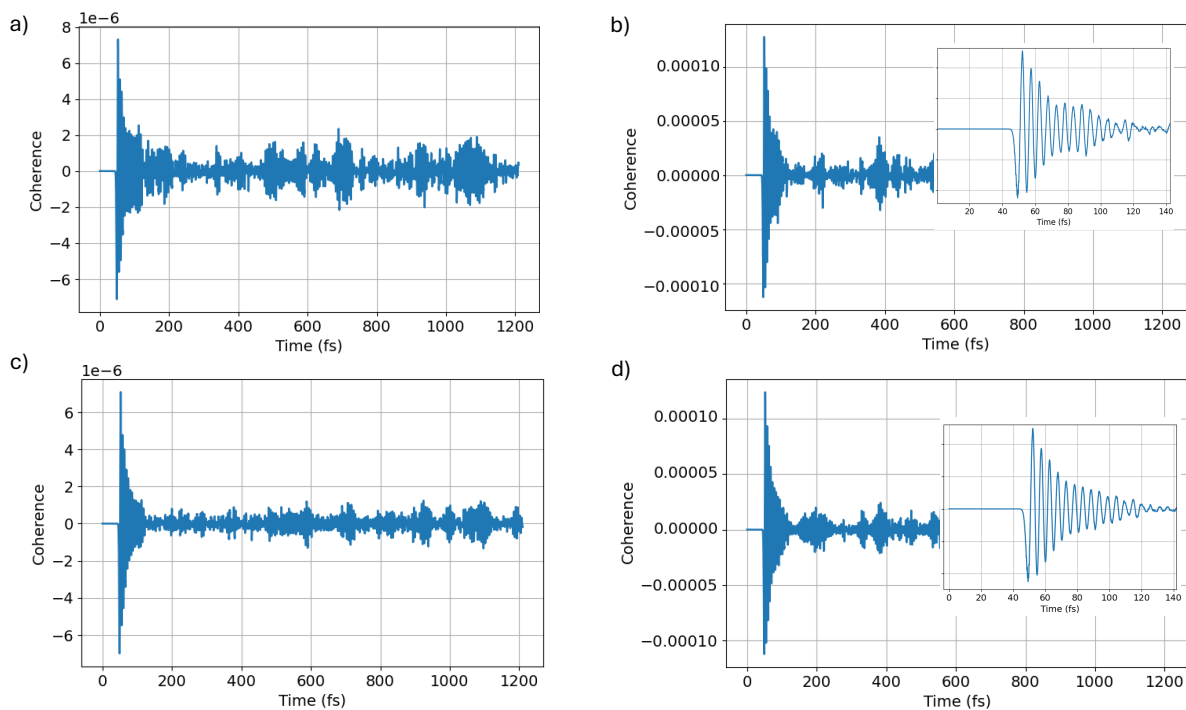


Figure S5: Evolution of the coherences as a function of time, when the system is interacting with a single pulse and pure electronic dephasing is included. a) Coherence between states 12-15, averaged over 100 trajectories. b) Coherence between states 13-15, averaged over 100 trajectories. c) Coherence between states 12-15, averaged over 200 trajectories. d) Coherence between states 13-15, averaged over 200 trajectories. In panel b) and b) the insight shows a magnification of the first 100 fs after the interaction with the pulse.