Supporting Information

Charge Transport Network Dynamics in Molecular Aggregates

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Simulation Box Protocol

Since this manuscript is primarily concerned with network methodological development for analyzing amorphous systems, our simulation procedure is not overly concerned with identifying the lowest energy non-crystalline morphology possible. If this were the case, a longer (~microseconds) and more gradual morphology simulation would be required. However, for the purposes of our short-time electronic dynamics (~20 ps), the following procedure is sufficient for generating a morphology required for electronic structure analysis.

Disordered PDI

1. A cubic box of length 100 Angstroms was randomly packed with 64 PDI molecules possessing random locations and orientations.
2. A force minimization is done in combination with a box compression to increase the density of the initially diffusely packed box, while simultaneously removing any bad contacts. This is followed by a brief (<20 ps) NVE run to reinitialize the packing of the box.
3. Another force minimization with box compression is performed to further compress the box to within a factor of 2 of the expected density. This is once again followed by a brief 20 ps NVE re-initialization of the box geometry.

4. A 100 ps NPT heating run is performed at 1 atm from 10K to 550K, the annealing temperature. The velocity distribution is randomly initialized according to the Boltzmann distribution.
5. A 10 ns NPT annealing run at 550K
6. A 10 ns NPT cooling run from 550K to 298 K.
7. A 10 ns NPT equilibration run at 298K.
8. A 1 ns NVT equilibration run at the average box volume of the previous 10 ns run.
9. A 100 ps NVE equilibration.
10. A 20 ps NVE sampling run, where the data in the manuscript is taken at 10 fs intervals.

Disordered bBDT(TDPP)$_2$

11. A cubic box of length 100 Angstroms was randomly packed with 100 BTI molecules possessing random locations and orientations.
12. A force minimization is done in combination with a box compression to increase the density of the initially diffusely packed box, while simultaneously removing any bad contacts. This is followed by a brief (<20 ps) NVE run to reinitialize the packing of the box.
13. Another force minimization with box compression is performed to further compress the box to within a factor of 2 of the expected density. This is once again followed by a brief 20 ps NVE re-initialization of the box geometry.
14. A 100 ps NPT heating run is performed at 1 atm from 10K to 550K, the annealing temperature. The velocity distribution is randomly initialized according to the Boltzmann distribution.
15. A 10 ns NPT annealing run at 550K
16. A 10 ns NPT cooling run from 550K to 298 K.
17. A 10 ns NPT equilibration run at 298K.
18. A 1 ns NVT equilibration run at the average box volume of the previous 10 ns run.
19. A 100 ps NVE equilibration.
20. A 20 ps NVE sampling run, where the data in the manuscript is taken at 10 fs intervals.

Crystalline bBDT(TDPP)$_2$

21. The unit cell of bBDT(TDPP)$_{2\,w}$ was repeated in a 5x5x2 block composed of 100 molecules, and a simulation cell corresponding to the appropriate unit cell was constructued.
22. A force minimization is done to remove bad contacts. This is followed by a brief (<20 ps) NVE run to reinitialize the packing of the box.
23. A 100 ps NPT heating run is performed at 1 atm from 10K to 550K, the annealing temperature. The velocity distribution is randomly initialized according to the Boltzmann distribution.

24. A 10 ns NPT annealing run at 550K
25. A 10 ns NPT cooling run from 550K to 298 K.
26. A 10 ns NPT equilibration run at 298K.
27. A 1 ns NVT equilibration run at the average box volume of the previous 10 ns run.
28. A 100 ps NVE equilibration.
29. A 20 ps NVE sampling run, where the data in the manuscript is taken at 10 fs intervals.

OPLS Force-Field Parameters

Since standard OPLS force-field parameters do not accurately describe the atomic geometry, atomic partial charges, and dihedral angles of PDI and bBDT(TDPP)$_2$, many of these values were parameterized by hand using quantum-chemical calculations. All bond distances and angle values were taken from the B3LYP/6-31G** geometry optimized structures. Atomic partial charges were determined by performing B3LYP/6-31G** CHelpG calculations on the geometry optimized structure. Improper dihedral angles were selected to rigidify the pi-system. The specific OPLS2009 parameters provided below are taken from the TINKER .prm files, as they are presented in an easily digestible format.

PDI

```
atom          907   110 N "PDI N1 class 35" 7 14.007 3
atom          908         111 C "PDI C2 class 3" 6 12.011 3
atom          909         112 C "PDI C3 class 38" 6 12.011 3
atom          910         113 C "PDI C4 class 38" 6 12.011 3
atom          911         114 C "PDI C5 class 38" 6 12.011          3
atom          912         115 C "PDI C6 class 38" 6 12.011          3
atom          913         116 C "PDI C7 class 38" 6 12.011 3
atom          914         117 C "PDI C8 class 38" 6 12.011 3
atom          915         118 O "PDI O9 class 4" 8 15.999 1
atom          916          13 C "PDI C10 class 13" 6 12.011 4
atom          917          39 H "PDI H11 class 39" 1 1.008 1
atom          918          39 H "PDI H12 class 39" 1 1.008 1
bond          110         111 490.00 1.409
bond          110          13 337.00 1.497
bond          111         112 400.00 1.484
bond          112         113 469.00 1.385
bond          113         114 469.00 1.399
bond          114         115 469.00 1.396
bond          115         116 469.00 1.431
bond          116         117 469.00 1.429
bond          111         118 570.00 1.225
bond          115         115 469.00 1.470
bond          112         117 469.00 1.414
bond          113          39 367.00 1.083
bond          114          39 367.00 1.083
angle         111          110         111  70.00  123.8
angle         111          110          13  50.00  117.0
angle         110          111         112  70.00  117.3
angle         110          111         118  80.00  121.4
angle         112          111         118  80.00  121.3
angle         111          112         113  85.00  119.3
angle         111          112         117  85.00  121.1
angle         113          112         117  63.00  119.6
```

```
angle       112     113     114  63.00  120.4
angle       112     113     39   35.00  118.7
angle       114     113     39   35.00  120.9
angle       113     114     115  63.00  121.9
angle       113     114     39   35.00  117.8
angle       115     114     39   35.00  120.2
angle       114     115     115  63.00  122.4
angle       114     115     116  63.00  118.5
angle       115     115     116  63.00  119.1
angle       115     116     115  63.00  121.8
angle       115     116     117  63.00  119.1
angle       112     117     112  63.00  119.3
angle       112     117     116  63.00  120.4
angle       110     13      13   80.00  111.7
angle       110     13      36   35.00  103.1
torsion     111     110     111  112   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     111     110     111  118   0.000 0.0 1    2.100 180.0 2   0.000 0.0 3
torsion      13     110     111  112   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion      13     110     111  118   0.000 0.0 1    2.100 180.0 2   0.000 0.0 3
#side-chain torsion
torsion     111     110     13   13    0.000 0.0 1   0.000 180.0 2   0.000 0.0 3
torsion     111     110     13   36      0.000 0.0 1    0.000 180.0 2   0.000 0.0 3
torsion     110     111     112  113   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     110     111     112  117   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     118     111     112  113   0.000 0.0 1    2.100 180.0 2   0.000 0.0 3
torsion     118     111     112  117   0.000 0.0 1    2.100 180.0 2   0.000 0.0 3
torsion     111     112     113  114   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     111     112     113  39      0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     117     112     113  39      0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     111     112     117  112   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     111     112     117  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     117     112     113  114   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     113     112     117  112   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     113     112     117  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     112     113     114  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     112     113     114  39      0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     39      113     114  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     39      113     113  39      0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     39      113     114  39      0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     113     114     115  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     113     114     115  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     39      114     115  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     39      114     115  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     114     115     115  114   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     114     115     115  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     114     115     116  117   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     116     115     115  116   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     114     115     116  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     115     115     116  115   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     115     115     116  117   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     115     116     117  112   0.000 0.0 1    7.250 180.0 2   0.000 0.0 3
torsion     110      13      13   13   0.845 0.0 1   -0.962 180.0 2   0.713 0.0 3
torsion     110      13      13   36   0.000 0.0 1    0.000 180.0 2   0.464 0.0 3
charge      907     -0.148
charge      908      0.534
charge      909     -0.270
charge      910      0.010
charge      911     -0.220
charge      912      0.060
charge      913      0.050
charge      914      0.210
charge      915     -0.480
charge      916      0.040
charge      917      0.120
charge      918      0.140
vdw         907     3.2500 0.1700
vdw         908     3.7500 0.1050
vdw         909     3.5500 0.0700
vdw         910     3.5500 0.0700
```

```
vdw          911          3.5500 0.0700
vdw          912          3.5500 0.0700
vdw          913          3.5500 0.0700
vdw          914          3.5500 0.0700
vdw          915          2.9600 0.2100
vdw          916    3.5000 0.0660
vdw          917    2.4200 0.0300
vdw          918    2.4200 0.0300
torsion      3  35  3  38   0.000 0.0 1          7.250 180.0 2          0.000 0.0 3
torsion      3  35  3  4                0.000      0.0 1         7.250 180.0 2          0.000 0.0 3
torsion      13  35  3  38              0.000      0.0 1         0.000 180.0 2          0.000 0.0 3
imptors  111 112 113 114  0.000              0.0 1         0.001 180.0 2          0.000 0.0 3
imptors      0   0 111 118  0.000 0.0 1   7.250 180.0 2    0.000 0.0 3
```

bBDT(TDPP)$_2$

```
atom         907  101 S "Thiophene1 S" 16 32.060 2
atom         908        102 Cd "Thiophene1 C2" 6 12.011 3
atom         909        103 CS "Thiophene1 C3" 6 12.011 3
atom         910        104 CS "Thiophene1 C4" 6 12.011 3
atom         911        105 Cd "Thiophene1 C5" 6 12.011 3
atom         912        106 HA "Thiophene1 H1" 1 1.008 1
charge       907  0.03
charge       908 -0.18
charge       909 -0.17
charge       910       -0.12
charge       911  0.03
charge       912  0.16
vdw          907  3.5500 0.2500
vdw          908  3.5500 0.0700
vdw          909        3.5500     0.0700
vdw          910  3.5500           0.0700
vdw          911        3.5500     0.0700
vdw          912  2.4200           0.0300
atom         913 107 NA "diKetoPyrrole N" 7 14.007 3
atom         914 108 CW "diKetoPyrrole C2O" 6 12.011 3
atom         915       109 CW "diKetoPyrrole C3" 6 12.011 3
atom         916       110 CN "diKetoPyrrole C4" 6 12.011 3
atom         917       111 O "diKetoPyrrole C=O" 8 15.999 1
atom         918        13 CT "diKetoPyrrole SC-CH2" 6 12.011 4
charge       913 -0.21
charge       914  0.50
charge       915  0.12
charge       916 -0.25
charge       917 -0.50
charge       918        0.10
vdw          913        3.2500 0.1700
vdw          914  3.5500 0.0700
vdw          915  3.5500           0.0700
vdw          916        3.5500     0.0700
```

```
vdw         917       2.9600     0.2100
vdw         918       3.5500     0.0700
atom        919       101 S "Thiophene2 S" 16 32.060 2
atom        920       102 Cd "Thiophene2 C2" 6 12.011 3
atom        921       103 CS "Thiophene2 C3" 6 12.011 3
atom        922       104 CS "Thiophene2 C4" 6 12.011 3
atom        923       105 Cd "Thiophene2 C5" 6 12.011 3
atom        924       106 HA    "Thiophene2 H1" 1  1.008 1
charge      919       -0.10
charge      920  0.12
charge      921 -0.26
charge      922 -0.15
charge      923  0.13
charge      924  0.16
vdw         919  3.5500 0.2500
vdw         920  3.5500 0.0700
vdw         921  3.5500 0.0700
vdw         922  3.5500 0.0700
vdw         923  3.5500 0.0700
vdw         924  2.4200 0.0300
atom        925  112 S "BDT S" 16 32.060 2
atom        926       113 C "BDT C1" 6 12.011 3
atom        927       114 C "BDT C2" 6 12.011 3
atom        928  115 C "BDT C3" 6 12.011 3
atom        929       116 C "BDT C4" 6 12.011 3
atom        930       117 C "BDT C5" 6 12.011 3
atom        931       118 C "BDT SC C6" 6 12.011 4
atom        932       119 HA "BDT H1" 1 1.008 1
charge      925       -0.03
charge      926       -0.10
charge      927       -0.120001
charge      928  0.000001
charge      929  0.05
charge      930       -0.10
charge      931  0.10
charge      932  0.19
vdw         925  3.5500 0.2500
vdw         926       3.5500     0.0700
vdw         927       3.5500 0.0700
vdw         928       3.5500     0.0700
vdw         929       3.5500     0.0700
vdw         930       3.5500     0.0700
vdw         931       3.5500     0.0700
vdw         932       2.4200     0.0300
#thio1
bond        101  102  250.00 1.727
```

```
bond        102     103  546.00 1.371
bond        103     104  546.00 1.416
bond        104     105  546.00 1.389
bond        105     101  250.00 1.764
bond        102     106  367.00 1.08
bond        103     106  367.00 1.08
bond        104     106  367.00 1.08
bond        109     102  546.00 1.440
angle       102     101  105  74.00  91.8
angle       101     102  103  70.00 111.9
angle       101     102  106  35.00 119.6
angle       103     102  106  35.00 128.5
angle       102     103  104  70.00 113.1
angle       102     103  106  35.00 123.4
angle       104     103  106  35.00 123.6
angle       103     104  105  70.00 113.5
angle       103     104  106  35.00 124.6
angle       105     104  106  35.00 121.9
angle       101     105  104  70.00 109.7
#DPP
bond        105  109  546.00 1.442
bond        107     108  427.00 1.434
bond        107     109  427.00 1.395
bond        108     110  546.00 1.445
bond        109     110  546.00 1.398
bond        110     110  546.00 1.420
bond        108     111  570.00 1.230
bond        107      13  337.00 1.461
angle       101     105  109  70.00 105.9
angle       104     105  109  70.00 124.4
angle       108     107  109  64.00 111.5
angle       108     107   13  70.00 118.4
angle       109     107   13  70.00 130.1
angle       107     108  110  70.00 104.2
angle       107     108  111  80.00 122.2
angle       110     108  111  80.00 133.6
angle       107     109  102  70.00 127.1
angle       107     109  110  70.00 106.6
angle       110     109  102  70.00 126.3
angle       105     109  107  70.00 127.0
angle       105     109  110  70.00 126.3
angle       108     110  109  70.00 142.3
angle       108     110  110  70.00 108.2
angle       109     110  110  70.00 109.5
angle       107      13   13  58.35 113.0
angle       107      13   36  35.00 107.0
```

```
angle          109        102  101  70.00 125.9
angle          109        102  103  70.00 125.8
#BDT
bond           105        113  546.00 1.441
bond           112        113  250.00 1.771
bond           113        114  546.00 1.373
bond           114        115  546.00 1.432
bond           115        116  546.00 1.414
bond           116        112  250.00 1.751
bond           116        116  546.00 1.393
bond           115        117  546.00 1.432
bond           117        118  469.00 1.518
bond           117        117  546.00 1.395
bond           114        119  367.00 1.080
bond           118         13  367.00 1.080
bond           118         36  268.00 1.529
angle          101        105  113  70.00 120.9
angle          104        105  113  70.00 128.8
angle          113        112  116  74.00 90.9
angle          105        113  112  70.00 119.9
angle          105        113  114  70.00 128.7
angle          112        113  114  70.00 111.3
angle          113        114  115  70.00 114.5
angle          113        114  119  35.00 125.9
angle          115        114  119  35.00 123.6
angle          114        115  116  70.00 111.0
angle          114        115  117  70.00 128.0
angle          116        115  117  70.00 121.0
angle          112        116  115  70.00 112.3
angle          112        116  116  70.00 128.3
angle          115        116  116  70.00 119.7
angle          115        117  117  70.00 119.2
angle          115        117  118  70.00 117.6
angle          117        117  118  70.00 123.2
angle          117        118   36  35.00 107.0
angle           13        118   36  35.00 107.0
angle          118   13   13  58.35 113.0
angle          118         13   36  35.00 107.0
angle          117        117   13  70.00 123.2
angle           36        118   36  33.00 107.8
angle          117  118   13  58.35 112.7
#torsion
torsion         0  103  104    0      0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
torsion         0  102  103    0      0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
torsion         0  101  102    0      0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
torsion         0  101  105    0      0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
```

```
torsion         0  104  105    0       0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
torsion         0  101  102    0       0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000 180.0 4
#DIHEDRAL THIO 1
torsion         101        105  109  107          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3  0.000          180.0 4
torsion         104        105  109  107          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3  0.000          180.0 4
#torsion        104        105  109  110          -2.017 0.0 1  8.394 180.0 2 -1.725 0.0 3  0.151          180.0 4
torsion     104  105  109  110  -1.906 0.0 1 8.514 180.0 2 -1.613 0.0 3  0.271 180.0 4
torsion         101        105  109  110          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3  0.000          180.0 4
#
torsion         0          107  108    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000          180.0 4
torsion         0  107  109    0       0.000 0.0 1  7.250 180.0 2  0.000 0.0 3  0.000          180.0 4
torsion         0          107   13   13          1.000 0.0 1 -0.350 180.0 2  0.000 0.0 3  0.000          180.0 4
torsion         0   13   13  107       0.000 0.0 1  0.000 180.0 2  0.462 0.0 3  0.000          180.0 4
torsion         0          107   13   36   0.000 0.0 1  0.000 180.0 2  0.000 0.0 3  0.000       180.0 4
torsion         0          108  110    0          0.000 0.0 1 10.750 180.0 2  0.000 0.0 3
torsion         0          110  110    0          0.000 0.0 1 10.750 180.0 2  0.000 0.0 3
torsion         0          109  110    0          0.000 0.0 1 10.750 180.0 2  0.000 0.0 3
#DIHEDRAL THIO 2
torsion         107        109  102  101   0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         107        109  102  103          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         110        109  102  101          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         110        109  102  103          -2.100 0.0 1  8.536 180.0 2 -1.840 0.0 3 -0.009 180.0 4
#
#DIHEDRAL 3 THIO-BDT
torsion         101        105  113  112          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         101        105  113  114          0.133 0.0 1  2.754 180.0 2 -0.195 0.0 3 -1.011 180.0 4
torsion         104        105  113  112          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         104        105  113  114          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
#
torsion         0  112  113    0   0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          112  116    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          113  114    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          114  115    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          115  116    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          115  117    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          116  116    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          117  117    0          0.000 0.0 1  7.250 180.0 2  0.000 0.0 3
torsion         0          117  118   13          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         0          117  118   36          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion         0          118   13   13          1.000 0.0 1 -0.350 180.0 2  0.000 0.0 3
torsion         118         13   13   13          1.300 0.0 1 -0.050 180.0 2  0.000 0.0 3
torsion         118         13   13   36          0.000 0.0 1  0.000 180.0 2  0.300 0.0 3
torsion          36        118   13   36          0.000 0.0 1  0.000 180.0 2  0.300 0.0 3
torsion         117        118   13   36          0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
#these parameters are just for refitting purposes
bond  109  106 367.0 1.08
```

```
bond  116   13 545.0 1.51
bond  113  106 367.0 1.08
bond  117  13  545.0 1.51
angle 110  109  106  35.00 130.4
angle 106  109          107  35.00 120.9
angle 115  117          13   70.00 120.5
angle 114  113          106  35.00 127.7
angle 106  113          112  70.00 119.3
angle 117   13          36  35.00 112.5
torsion       115  117  13  36  0.000 0.0 1  0.000 180.0 2  0.000 0.0 3
torsion       117  117  13          36  0.000 0.0 1          0.000 180.0 2          0.000 0.0 3
imptors       0    0 110   0  0.000 0.0 1          5.000 180.0 2          0.000 0.0 3
```

Example Input File for Disordered PDI

Control file

```
# ----------------- Init Section -----------------

include "box_of_PDI_label_min.in.init"

# ----------------- Atom Definition Section ----------------------

read_data "box_of_PDI_label_min.data"

# ----------------- Setting Section -----------------

include "box_of_PDI_label_min.in.settings"

# ----------------- Run Section -----------------

#USING RATTLE TO GET TIMESTEP OF 2
thermo 1000
thermo_style custom step dt temp press vol etotal ke pe ebond eangle edihed eimp evdwl ecoul elong density
neigh_modify delay 0 every 1 check yes page 1000000 one 100000
run_style verlet

#QUICK COMPRESS TO CLEAN UP THE SIMULATION BOX
dump 1 all atom 1000 min.dump
fix 1 all box/relax iso 10000000.0 vmax 0.01
minimize 1.0e-6 1.0e-6 20000 200000
unfix 1
undump 1

#NVE EQUILIBRATION RUN (20 ps)
timestep 1
fix 2 all nve/limit 0.01
run 20000
unfix 2

#Second Compression Minimization
fix 3 all box/relax iso 1000000.0 vmax 0.01
minimize 1.0e-6 1.0e-6 20000 200000
unfix 3

#Second NVE Equil
timestep 1
fix 4 all nve/limit 0.01
run 20000
unfix 4
```

```
#Heating up
timestep 2
neigh_modify delay 5 every 1 check yes page 1000000 one 100000
fix 5 all npt temp 10.0 550.0 100.0 iso 1.0 1.0 1000
fix 15 all rattle 0.0001 20 0 m 1.008
run 50000
unfix 15
unfix 5

#ANNEALING RUN (550K for 2 ns)
fix 6 all npt temp 550.0 550.0 100.0 iso 1.0 1.0 1000
fix 16 all rattle 0.0001 20 0 m 1.008
run 5000000
unfix 16
unfix 6

#COOLING (550K - 298K over 100 ps)
fix 7 all npt temp 550.0 298.0 100 iso 1.0 1.0 1000
fix 17 all rattle 0.0001 20 0 m 1.008
run 5000000
unfix 17
unfix 7

#SIMULATION (nPT for 6 ns; 1 ns snapshots)
dump 8 all atom 1000000 equil.dump
fix 8 all npt temp 298.0 298.0 100 iso 1.0 1.0 1000
fix 18 all rattle 0.0001 20 0 m 1.008
run 5000000
unfix 18
unfix 8
undump 8

#NVT Run (nVT for 2 ns)
fix 200 all nvt temp 298.0 298.0 100
fix 201 all rattle 0.0001 20 0 m 1.008
run 500000
unfix 201
unfix 200

#NVE Equil
timestep 1
fix 700 all nve
run 50000
unfix 700

#SAMPLING (NVE for 20 ps; 10 fs snapshots)
timestep 1
dump 100 all atom 10 sample.dump
fix 100 all nve
run 20000
unfix 100
undump 100

#END OF RUN SECTION #
```

.init file

```
units          real          # angstroms,kCal/mol,Daltons,Kelvin
atom_style     full          # select column format for Atoms section
pair_style  lj/cut/coul/long 10.0 10.0       # cutoffLJ cutoff_coulomb
bond_style     harmonic      #parameters needed: k_bond, r0
angle_style    harmonic      #parameters needed: k_theta, theta0
dihedral_style opls
improper_style cvff
kspace_style   pppm 0.0001   # long-range electrostatics sum method
pair_modify mix arithmetic   #using Lorenz-Berthelot mixing rules
```

Simulation Details

The data of Figure 3 was fit using the Optimize.curve_fit routine in Scipy. This routine employ a non-linear least squares to fit a function, f, to the data. The data was fit to the form:

TCF(t) = A1*exp(-b1*t) + A2*exp(-b2*t)

The optimal values of the fitting parameters were the following:

Disordered PDI

A1, A2, b1, b2
```
[ 0.49960115  0.48670783  0.00526324  0.03193688]
```

and the covariance matrix was:

```
[[  9.02869987e-05  -7.80417345e-05   6.92676478e-07   8.72171357e-06]
 [ -7.80417345e-05   9.45332078e-05  -6.16078009e-07  -5.73017840e-06]
 [  6.92676478e-07  -6.16078009e-07   6.30505437e-09   6.06600711e-08]
 [  8.72171357e-06  -5.73017840e-06   6.06600711e-08   1.19407013e-06]]
```

Disordered bBDT(TDPP)$_2$

A1, A2, b1, b2
```
[ 0.4235485   0.57582765  0.18934241  0.01063638]
```

and the covariance matrix was:

```
[[  2.11630386e-04  -1.10262518e-04  -1.17334933e-04  -1.89822556e-06]
 [ -1.10262518e-04   1.11650354e-04   1.42336942e-04   1.91805797e-06]
 [ -1.17334933e-04   1.42336942e-04   4.19904609e-04   2.24795900e-06]
 [ -1.89822556e-06   1.91805797e-06   2.24795900e-06   4.80374715e-08]]
```

Crystalline bBDT(TDPP)$_2$

A1, A2, b1, b2
```
[ 0.80770858  0.14074453  0.01317332  0.00311807]
```

and the covariance matrix was:

```
[[  3.77322746e-04  -3.76912356e-04  -6.12503629e-06  -5.15227374e-06]
 [ -3.76912356e-04   4.25922587e-04   7.81860648e-06   5.69374574e-06]
 [ -6.12503629e-06   7.81860648e-06   1.76896233e-07   9.63502632e-08]
 [ -5.15227374e-06   5.69374574e-06   9.63502632e-08   8.46924134e-08]]
```

Local TCF Fourier Transform

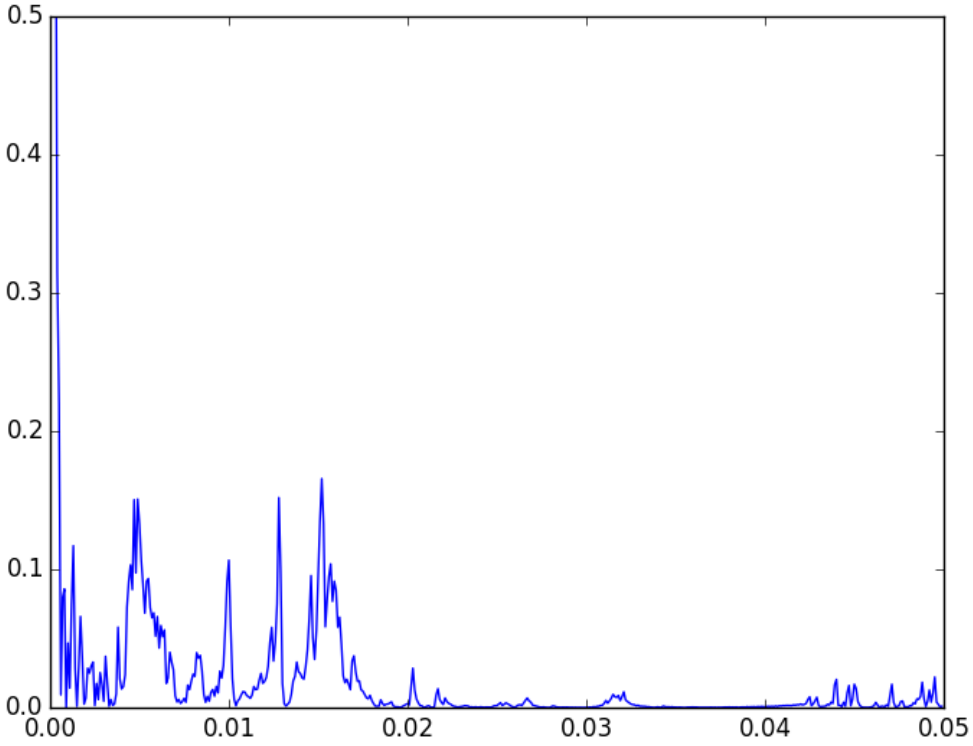

Figure S1 – Fourier spectrum of the ensemble averaged time-correlation function of the intermolecular electronic coupling between PDI LUMO's.

TCF of Matrix Spectral Norm

To quantify how the self-similarity of the graph representing the charge transport network of the molecular aggregate evolves as a function of time, we compute a time-correlation function of the adjacency matrix spectral norm defined below. TCF reveals a decorrelation timescale of ~100 fs, in agreement with the qualitative assignments derived from visual inspection of Figure 2 in the manuscript.

$$C_{ij}(\tau) = \lim_{T\to\infty} \int_0^T \frac{SpecNorm[A(t) - A(t+\tau)]}{T} dt$$

Where Norm represents the spectral norm, and A is the adjacency matrix at time t. The Spectral Norm TCF for all 5 trajectories are shown below.

PDI - Trajectory 1

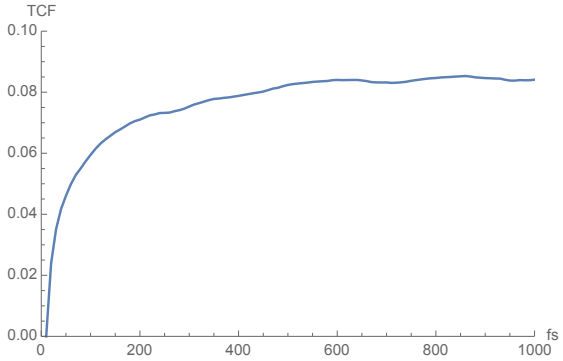

Figure S2 – TCF of the spectral norm of the adjacency matrix for PDI trajectory # 1.

PDI - Trajectory 2

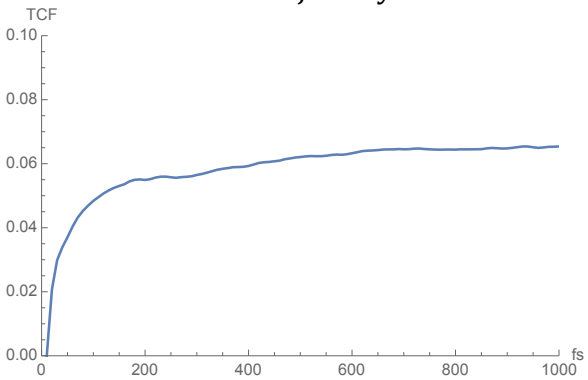

Figure S3 – TCF of the spectral norm of the adjacency matrix for PDI trajectory # 2.

PDI - Trajectory 3

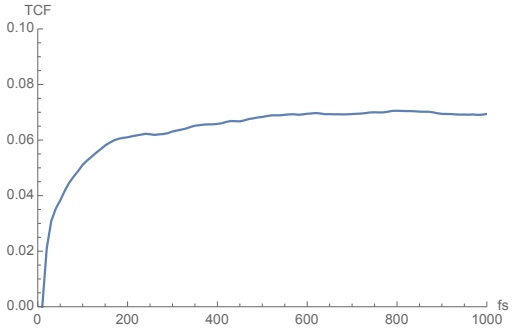

Figure S4 – TCF of the spectral norm of the adjacency matrix for PDI trajectory # 3.

PDI - Trajectory 4

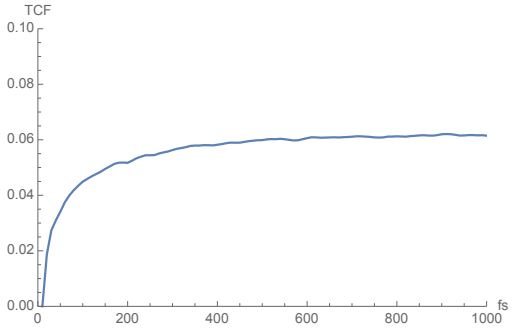

Figure S5 – TCF of the spectral norm of the adjacency matrix for PDI trajectory # 4.

PDI - Trajectory 5

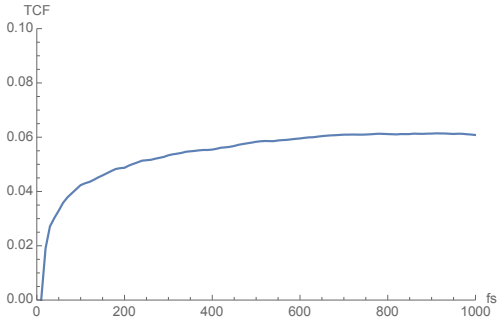

Figure S6 – TCF of the spectral norm of the adjacency matrix for PDI trajectory # 5.

Correlations between Kirchoff Index and Zero-Field Mobilities Using a Kinetic Monte Carlo Model

While the correlation between the Kirchoff Index and experimental mobilities or diffusion constants has only been qualitative in previous work (Jackson – JPCL – 2015 – 1:1018-1021), here we use the same geometric model as in our previous work to generate example morphologies. Details on the description of the method for generating morphologies and the lattice model used can be found in our previous JPCL manuscript. These morphologies are then used to generate a realistic ensemble of charge transport networks defined by an adjacency matrix with elements corresponding to intermolecular couplings. These intermolecular couplings are used as inputs into Fermi's Golden Rule to derive rate constants of hopping between sites in the morphology. Fermi's Golden Rule is used for comparison to our Kirchoff Index using the Adjacency Matrix definition of the absolute value of the electronic coupling because both terms ignore energetic disorder, and infer rate constants only from the magnitude of the intermolecular electronic coupling. In a system in which energetic disorder were consider, one would use a Marcus-Jortner-Levich type expression, and a definition of the adjacency matrix which incorporates energetic disorder.

Using these rates, a master equation between all sites in the system is established, and approximately solved using a kinetic monte carlo (KMC) approach. Simulations are run for 10 ns, and the location of the charge on the lattice is computed at the beginning and end of the trajectory. A cubic lattice containing 3,375 sites is used. A maximum coupling of 0.025 eV is allowed between nearest neighbor sites in our model, and these values vary continuously depending on the relative orientations of nearest neighbor molecules (again, see previous JPCL work for more details on the model).

Finally, the mean-squared hopping distance (delta r)^2 can be related to the zero-field mobility or a diffusion constant via an Einstein-like relation. Consequently, comparison of the Kirchoff index of our example networks to the KMC derived (delta r)^2 allows for a rigorous test of the relationship between the Kirchoff index and charge mobilities/diffusion constants. The results of these simulations are shown in Figure S7, where a clearly positive, monotonic relationship between charge mobility and the kirchoff index is observed for the 250 independently sampled morphologies, providing proof of the utility of the Kirchoff index as a qualitative metric of charge mobilities.

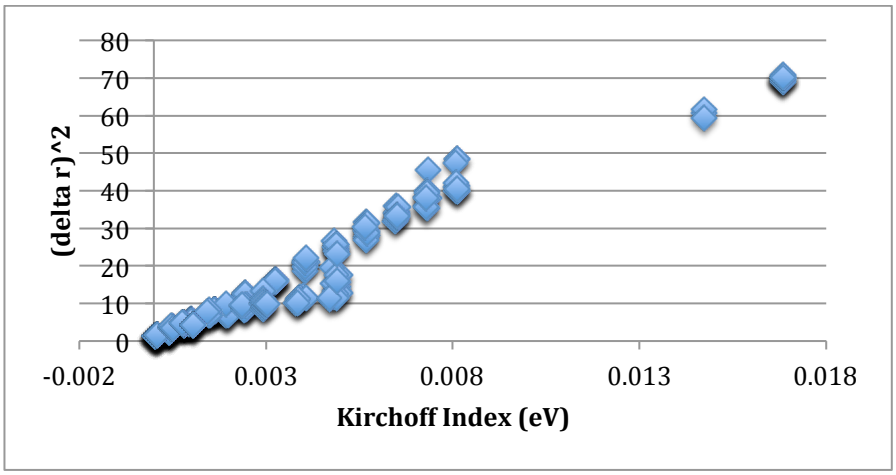

Figure S7 – Plot of KMC derived mean-squared displacement of charge vs network's Kirchoff Index. Units of delta r are in terms of lattice units.