Supporting Information

Diversifying Peripheral Aromatic Units of Pyrrolo[3,2-*b*]pyrrole-Containing Conjugated Polymers and the Resulting Optoelectronic Properties

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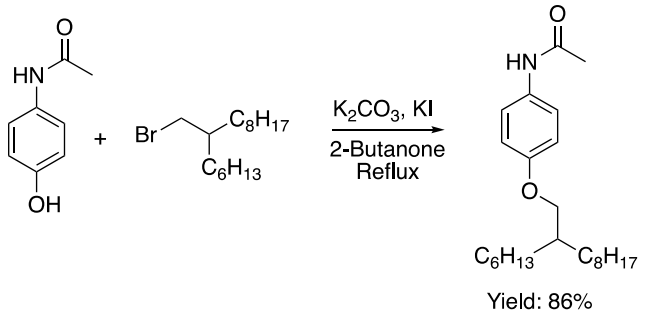
Materials and Methods

All materials used in synthetic protocols were purchased from commercial sources and used as received unless otherwise stated. Anhydrous acetonitrile (ACN) and toluene were obtained from a Pure Process Technology GC-SPS-7 Glass Contour 800L Solvent Purification System stored under argon (Ar) and degassed with Ar for 15 min. before use. All column chromatography purifications used 60 Å silica gel (200-400 mesh). ^{1}H NMR and ^{13}C NMR spectra were collected on a Bruker Advance III HD 400 MHz NMR spectrometer with nominal concentrations of 5 mg/mL in $CDCl_3$. Peaks are referenced to the residual $CHCl_3$ peak (^{1}H: δ = 7.26 ppm; ^{13}C: δ = 77.23 ppm). Polymer molecular weights were estimated via size exclusion chromatography (SEC) using a Tosoh EcoSEC HLC-8320 or an Agilent 1260 Infinity II system operated at 35 °C while using chloroform as the eluent. Polymer solutions (~5 mg/mL in $CHCl_3$) were prepared and filtered through a PTFE 0.45 μm filter prior to injection. Thermogravimetric analyses (TGA) were performed using a TA Instruments Discovery TGA550 TGA thermogravimetric analyzer in platinum pans under N_2. A scan used a ramp from room temperature to 800 °C at a heating rate of

10 °C/min. Differential scanning calorimetry (DSC) measurements were made using a TA instruments Discovery DSC250 DSC with a heat-cool-heat cycle from -25 °C to 250 °C at a heating rate of 5 °C/min. Optical absorbance spectra of solutions and films were acquired using a Varian Cary 5000 Scan dual-beam UV−vis−near-IR spectrophotometer. Cyclic voltammetry (CV) measurements were performed with a CH Instruments Electrochemical workstation (CHI660D), using a glassy carbon electrode as the working electrode, an Ag/AgCl reference electrode (calibrated versus the Fc/Fc^+ redox couple, $E_{1/2}$ = 46 mV), and a Pt flag as the counter electrode. A 50 mV/s scan rate was used for all electrochemical measurements. An electrolyte solution of 0.5 M tetrabutylammonium hexafluorophosphate ($TBAPF_6$, 98%) in anhydrous ACN was used for electrochemical measurements. Glass slides used for film studies were cleaned first by sonicating for ~5 min in acetone followed by isopropanol (IPA) and rinsed with the respective solvent before being dried with compressed air. Polymer films were deposited onto the glass slides from ~5-20 mg/mL $CHCl_3$ solutions using an Ossila Spin Coater at 1800 rpm for 30 s. There are no hidden risks or hazards to declare for this work.

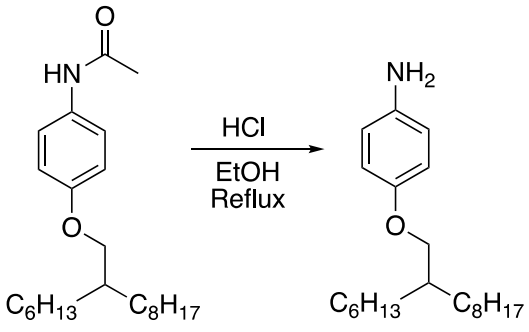
Synthesis and Characterization

General Procedure for Synthesizing Ether-Functionalized Anilines from Acetaminophen



Scheme S1. General synthesis of ether-functionalized acetaminophen with the corresponding alkyl bromides.

All ether-functionalized anilines were synthesized following a literature procedure reported by adopted by our group.[1,2] Acetaminophen (8.00 g, 53.0 mmol), potassium iodide (KI) (0.439 g, 3.00 mmol), and K_2CO_3 (18.3 g, 133 mmol) were added to a 250 mL three-neck round bottom flask equipped with a condenser and magnetic stir bar. The flask was sealed with three rubber septa and rendered inert via 3 vacuum/refill cycles with Ar. 2-Butanone (132 mL) was added to the flask for a reaction concentration of 1.2 M with respect to the acetaminophen and alkyl bromide. The mixture was lowered into a preheated oil bath set to 90 °C for 30 minutes. After heating for 30 min., 7-(bromomethyl)pentadecane (106 mmol, 2.0 eq) was added to the flask via syringe and the reaction was set to run overnight (~16-20 h). The following day, the reaction was removed from the oil bath and cooled to room temperature. The reaction was suspended in 150 mL of water before extraction with ethyl acetate (3×30 mL). The combined ethyl acetate layers were washed with water (3×100 mL), dried over anhydrous Na_2SO_4, and filtered. The organic phase was concentrated via rotary evaporation to yield an amber-yellow oil. The crude product was dried under vacuum overnight and used in the subsequent procedure without further purification.

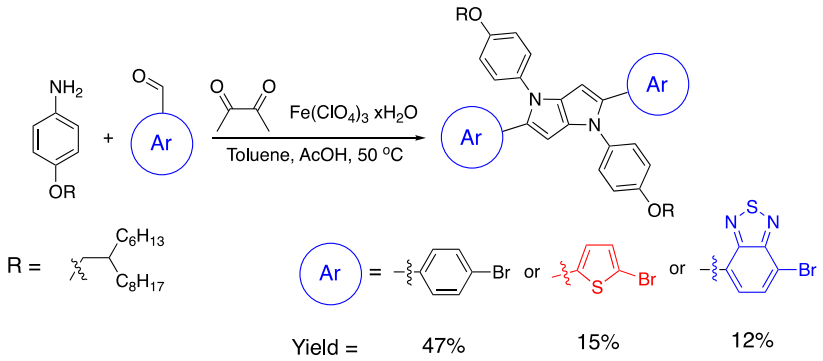


Scheme S2. Deprotection reaction for synthesizing ether-functionalized anilines via acetyl-group hydrolysis.

The ether-functionalized acetaminophen (20 mmol) was transferred into a 100 mL round bottom flask equipped with a magnetic stir bar and dissolved in 90% ethanol (48 mL). Concentrated HCl (18 mL) was then added to the reaction mixture dropwise and a condenser was attached. The reaction was lowered into a preheated oil bath set to 90 °C and the reaction was allowed to run overnight (~16-20 h). After stirring overnight, the reaction was removed from heat and cooled to room temperature. The reaction mixture was transferred to a beaker and diluted with ~ 100 mL of water. Aqueous NaOH (1.0 M) was added to the mixture until pH paper indicated the solution was neutralized. The desired product was extracted with DCM (3×50 mL), dried over anhydrous Na_2SO_4, and filtered. The organic phase was concentrated via rotary evaporation to yield an amber oil. The crude product was dried under vacuum overnight and used in subsequent procedures without further purification.

4-((2-hexyldecyl)oxy)-aniline: 4.25 g (64%). ^{1}H NMR (400 MHz, $CDCl_3$), δ= 0.90 (t, 6H), 1.30 (br m, 20H), 1.44 (m, 4H), 1.76 (m, 1H), 3.42 (br s, 2H), 3.77 (d, 2H), 6.67 (d, 2H), 6.76 (d, 2H). ^{13}C NMR (400 MHz, $CDCl_3$), δ: 14.12, 22.69, 26.83, 26.86, 29.34, 29.61, 29.72, 31.40, 31.88, 31.92, 38.08, 71.75, 115.71, 116.39, 139.72, 152.70. Structural characterization matched previously reported data.[1]

Synthesis of Functionalized DHPP Monomers via an Fe(III)-Catalyzed Multicomponent Reaction



Scheme S3. General synthesis of the brominated DHPP monomers via an Fe(III)-catalyzed multicomponent reaction.

The synthetic protocol for DHPPs was adopted from previous work reported by our group. Briefly, 4-((2-hexyldecyl)oxy)-aniline (8 mmol) and the corresponding aldehyde (8 mmol) were added to a solution of toluene (6 mL) and glacial acetic acid (6 mL) inside a 25 mL round bottom flask equipped with a magnetic stir bar. The reaction mixture was stirred for 1 h in an oil bath set to 50 ˚C. Once the initial heating time was completed, $Fe(ClO_4)_3 \cdot xH_2O$ (0.085 g) was added to the reaction flask, followed by 2,3-butanedione (0.35 mL, 4.00 mmol). After these additions, the reaction was allowed to stir at 50 ˚C overnight. The next day the reaction was removed from heat and allowed to cool to room temperature. Each monomer required slightly different purification protocols and are reported below. Following purification via washing or recrystallization, the solids were collected in a vial and dried under vacuum overnight. After structural analysis, each monomer was confirmed to be the desired product.

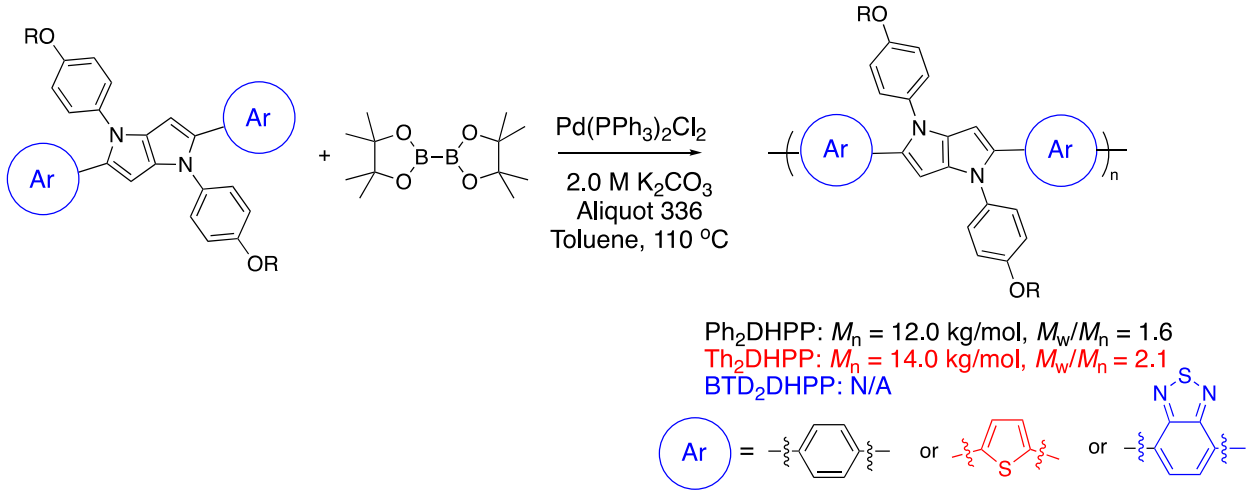
2,5-bis(4-bromophenyl)-1,4-bis(4-((2-hexyldecyl)oxy)phenyl)-1,4-dihydropyrrolo[3,2-*b*]pyrrole (*m*-Ph_2DHPP): The reaction precipitate was collected via vacuum filtration and washed with cold MeOH and acetone until a pale-yellow solid remained on the filter paper. Yield: 1.74 g (47%). ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.93 (br, 12H), 1.34 (br, 48 H), 1.81 (m, 2H), 3.87 (d, 4H), 6.34 (s, 2H), 6.93 (d, 4H), 7.10 (d, 4H), 7.19 (d, 4H), 7.34 (d, 4H). ^{13}C NMR (400 MHz, $CDCl_3$), δ: 14.12, 22.69, 26.87, 29.34, 29.61, 29.70, 30.03, 31.40, 31.87, 31.92, 38.03, 71.26, 93.86, 115.06, 119.95, 126.54, 129.44, 131.27, 132.09, 132.52, 132.60, 134.94, 157.65. Anal. calc'd for $C_{61}H_{82}Br_2N_2O_2$: C 70.78, H 7.98, Br 15.44, N 2.71, O 3.09. Found: C 71.17 H 8.07, N 2.70.

2,5-bis(5-bromothiophen-2-yl)-1,4-bis(4-((2-hexyldecyl)oxy)phenyl)-1,4-dihydropyrrolo[3,2-*b*]pyrrole (*m*-Th$_2$DHPP): After cooling to room temperature, the crude reaction mixture was precipitated dropwise into chilled MeOH. The precipitate was a brown sludge that was collected via vacuum filtration and washed with cold MeOH and acetone until a pale-yellow solid remained on the filter paper. Yield: 168 mg (15%). ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.92 (br, 10H), 1.32 (br, 50 H), 1.83 (q, 2H), 3.90 (d, 4H), 6.23 (s, 2H), 6.39 (d, 2H), 6.81 (d, 2H), 6.98 (dt, 4H, J = 8.0, 2.0 Hz), 7.28 (m, 4H, J = 8.0 Hz). ^{13}C NMR (400 MHz, $CDCl_3$), δ: 14.14, 22.71, 26.88, 26.91, 29.36, 29.62, 29.71, 30.05, 31.41, 31.88, 31.94, 38.04, 71.32, 93.08, 115.09, 124.43, 127.72, 129.98, 131.57, 158.56. Anal. calc'd for $C_{58}H_{80}Br_2N_2O_2S_2$: C 65.65, H 7.60, Br 15.06, N 2.64, O 3.02, S 6.04. Found: C 65.55 H 7.57, N 2.57, S 6.20.

7,7'-(1,4-bis(4-((2-hexyldecyl)oxy)phenyl)-1,4-dihydropyrrolo[3,2-*b*]pyrrole-2,5-diyl)bis(4-bromobenzo[*c*][1,2,5]thiadiazole) (*m*-BTD$_2$DHPP): After cooling to room temperature, the crude reaction mixture was precipitated dropwise into chilled MeOH. The precipitate was a dark sludge that was collected via vacuum filtration and washed with cold MeOH and acetone to remove a significant portion of unknown impurities. The solid was then transferred into an Erlenmeyer flask and recrystallized from ethyl acetate. The recrystallized product was collected via filtration and washed with chilled ethyl acetate to yield a dark-red solid. Yield: 278 mg (12%). ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.88 (br, 10H), 1.29 (br, 50H), 1.76 (t, 2H), 3.82 (d, 4H), 6.88 (dt, 4H, J = 8.0, 2.0 Hz), 6.98 (d, 2H, J = 8.0 Hz), 7.01 (s, 2H), 7.23 (dt, 4H, J = 8.0, 2.0 Hz), 7.61 (d, 2H, J = 8.0 Hz). ^{13}C NMR (400 MHz, $CDCl_3$), δ: 14.13, 22.69, 26.86, 26.90, 29.34, 29.61, 29.70, 30.03, 31.39, 31.87, 31.91, 38.02, 71.29, 77.22, 98.35, 110.86, 115.21, 126.59, 127.77, 131.83, 134.06,

152.85, 157.81. Anal. calc'd for $C_{61}H_{82}Br_2N_2O_2$: C 63.91, H 6.92, Br 15.44, N 7.21, O 3.09, S 5.50. Found: C 63.27 H 6.70, N 7.12, S 5.53.

General Polymerization Protocol



Scheme S4. Generic synthetic pathway for Suzuki polycondensations that produce DHPP copolymers.

The dihalogenated DHPP (1.0 molar equiv.), bis(pinacolato)diboron (1 molar equiv.), and 2 mol% of $Pd(PPh_3)Cl_2$ were added to a 10 mL, one-neck round bottom flask along with a Teflon stir bar. One drop of Aliquat 336 was subsequently added to the flask. A condenser was connected to the round bottom flask and sealed with a rubber septum. The flask was rendered inert via 3× vacuum/refill cycles with Ar before adding 2M K_2CO_3 (aq) and toluene in a 4:1 ratio via syringe. The reaction mixture was placed in an oil bath set to 110 ˚C and stirred overnight. The next day, the reaction mixture was cooled to room temperature before precipitation into ~ 200 mL of MeOH while vigorously stirring. The precipitate was collected in a Soxhlet thimble and then washed with MeOH and acetone to remove impurities and low molecular weight oligomers before extracting the desired polymer from the Soxhlet thimble with chloroform. The product was concentrated via

rotary evaporation and precipitated into ~ 200 mL of MeOH while stirring. The precipitate was allowed to stir for 1 h before collecting the product via vacuum filtration. The polymer was placed into a vial and allowed to dry under vacuum overnight. All three polymers were confirmed to be the expected products after structural analysis.

Ph_2DHPP: Yellow Solid. Yield: 80 mg (70%) ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.88 (br, 17H), 1.29 (t, 60H), 1.77 (t, 2H), 3.83 (d, 4H), 6.38 (s, 2H), 6.89 (m, 4H), 7.23 (m, 8H), 7.46 (m, 4H). Anal. calc'd for $C_{62}H_{86}N_2O_2$: C 83.54; H 9.73; N 3.14 Found: C 82.65; H 9.56; N 3.21. M_n = 12.0 kg/mol M_w/M_n = 1.6.

Th_2DHPP: Red Solid. Yield: 105.5 mg (77%) ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.88 (br, 15H), 1.30 (br, 80H), 1.81 (t, 2H), 3.86 (d, 4H), 6.24 (s, 2H), 6.37 (d, 2H), 6.77 (br, 2H), 6.95 (dt, 4H, J = 8.7, 3.0 Hz), 7.29 (m, 4H). Anal. calc'd for $C_{58}H_{82}N_2O_2S_2$: C 77.11; H 9.15; N 3.10; S 7.10 Found: C 76.65; H 8.88; N 3.00; S 7.14. M_n = 14.0 kg/mol M_w/M_n = 2.1.

BTD_2DHPP: Dark-Blue Solid. Yield: 48.5 mg (53%). *Yields were lower because an insoluble blue material remained in the Soxhlet thimble.* ^{1}H NMR (400 MHz, $CDCl_3$), δ: 0.87 (br, 15H), 1.28 (br, 57H), 1.78 (br, 2H), 3.84 (br, 4H), 6.90 (m, 4H), 7.14 (br, 2H), 7.34 (m, 4H), 8.26 (br, 2H). Anal. calc'd for $C_{62}H_{82}N_6O_2S_2$: C 73.91; H 8.20; N 8.34; S 6.36 Found: C 74.04; H 8.08; N 8.18; S 6.19. Product did not possess adequate solubility for molecular weight estimation.

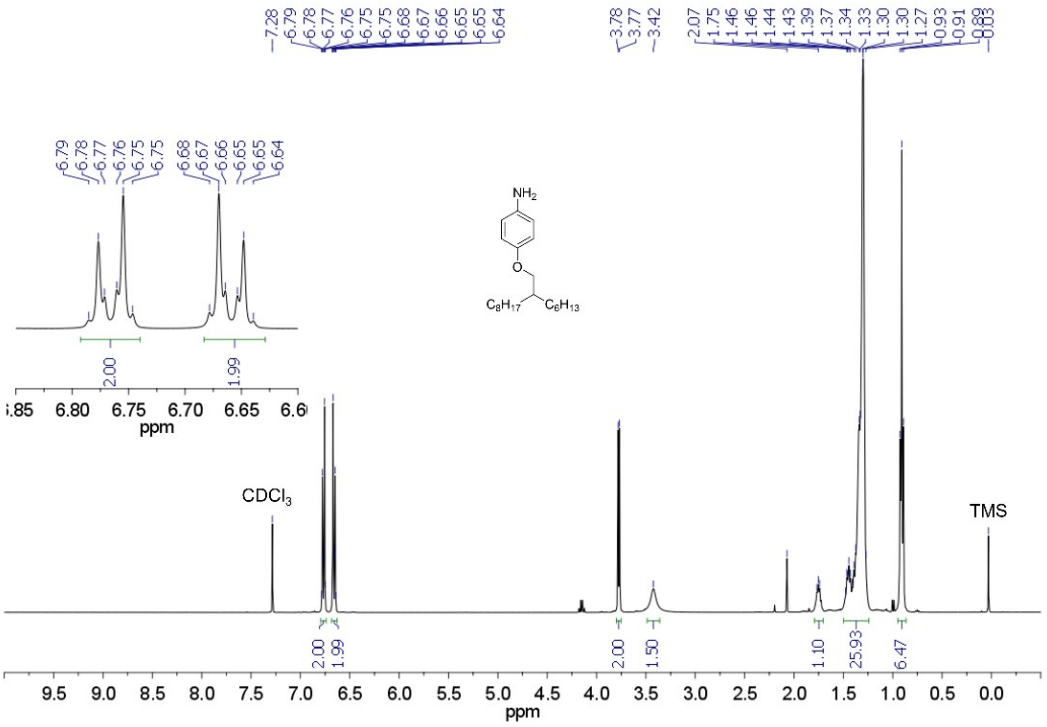


Figure S1. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of HxDecAniline.

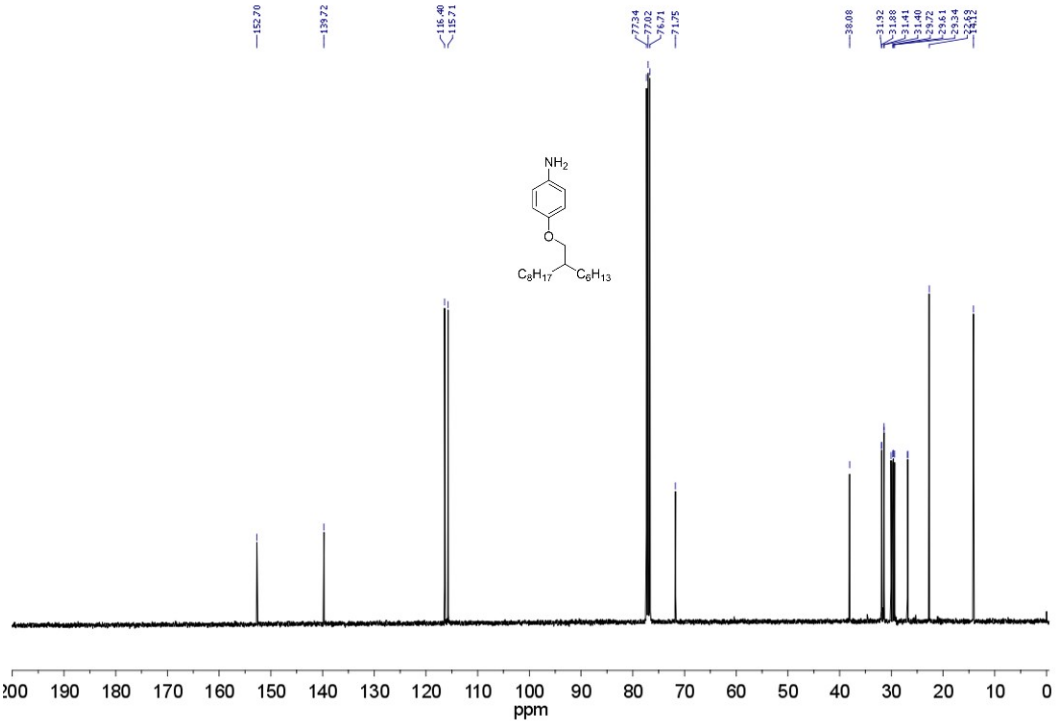


Figure S2. ^{13}C NMR (400 MHz, 25 °C, $CDCl_3$) of HxDecAniline.

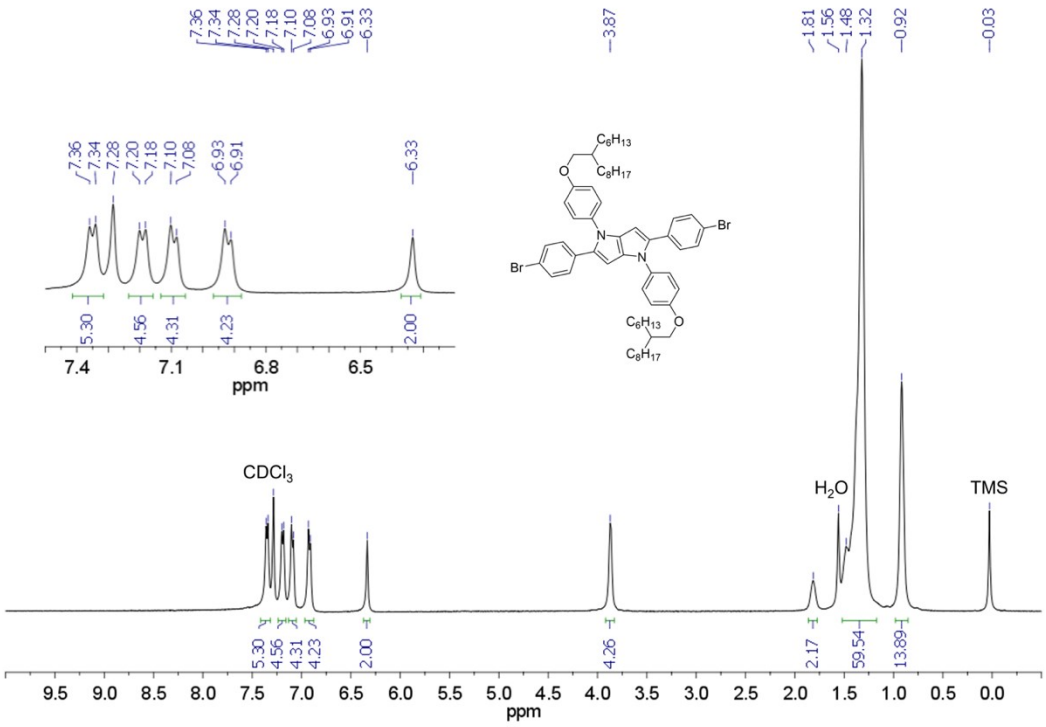


Figure S3. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-Ph_2DHPP.

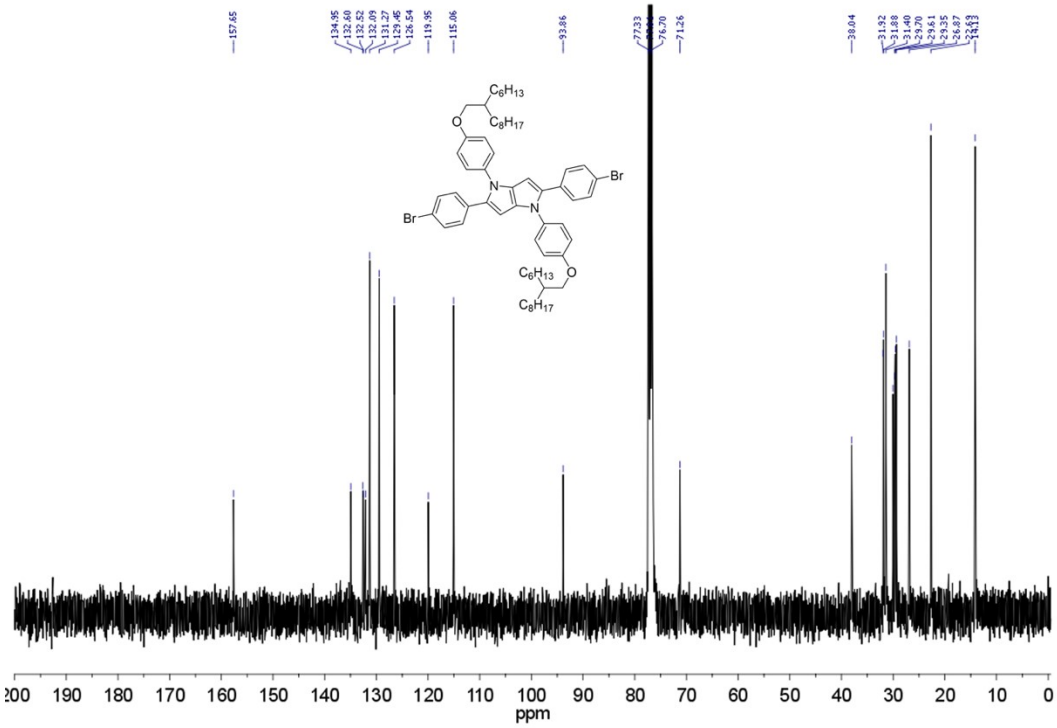


Figure S4. ^{13}C NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-Ph_2DHPP.

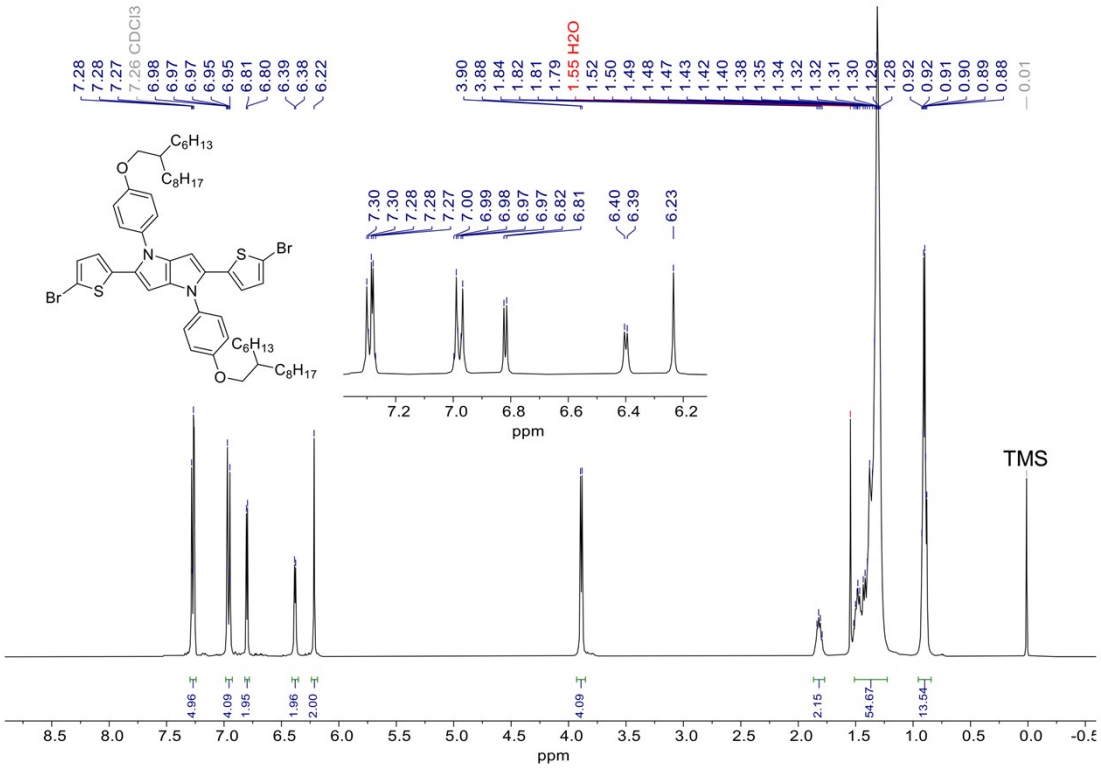


Figure S5. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-Th_2DHPP.

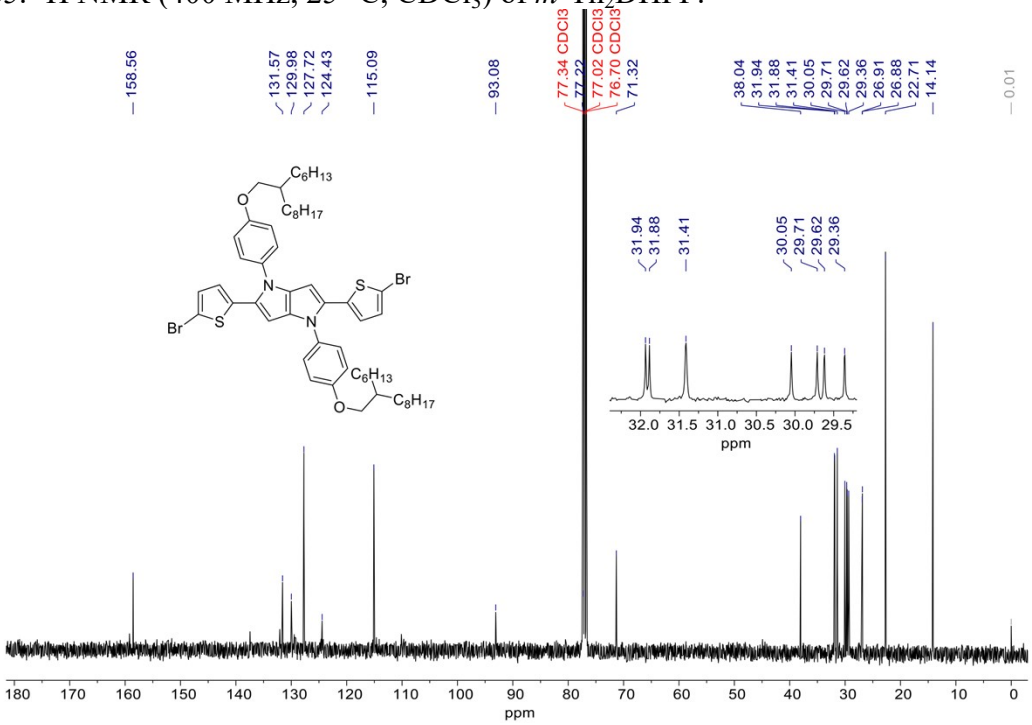


Figure S6. ^{13}C NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-Th_2DHPP.

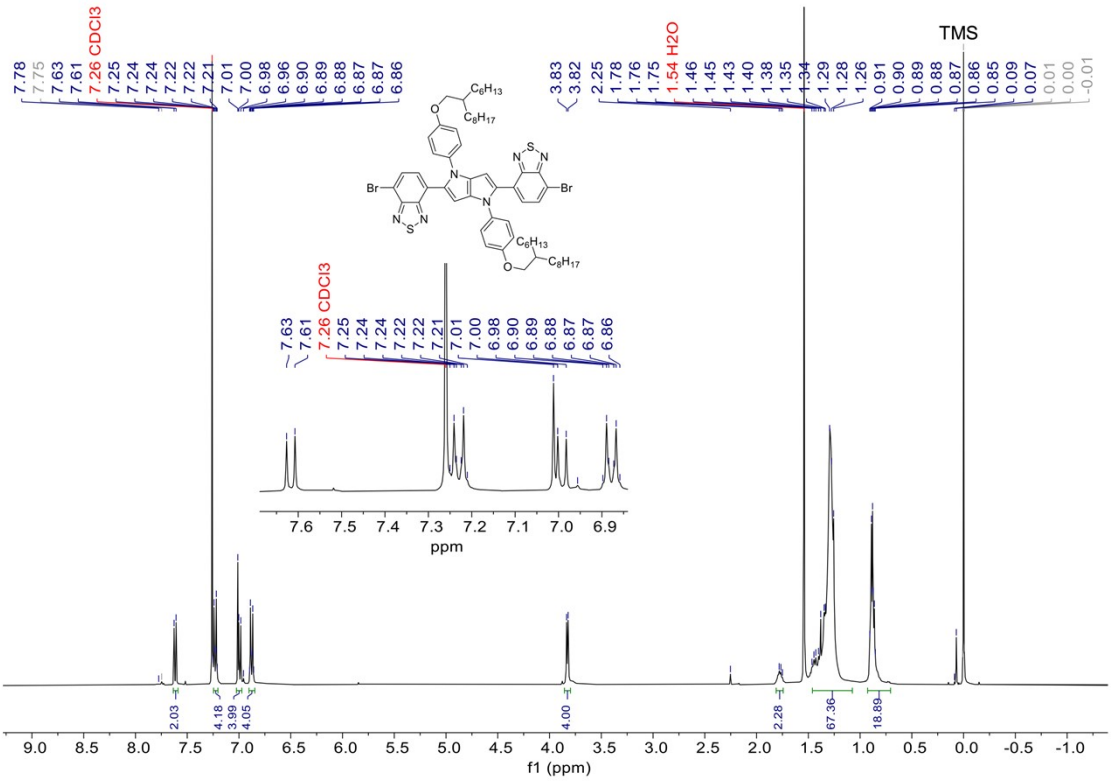


Figure S7. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-BTD_2DHPP.

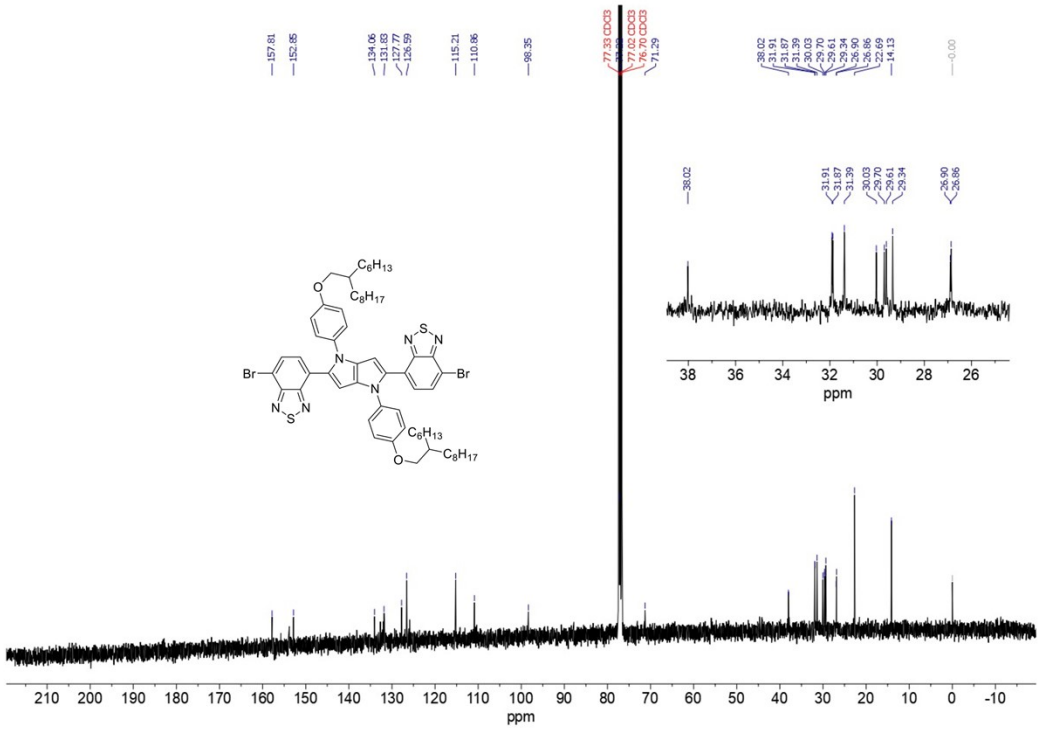


Figure S8. ^{13}C NMR (400 MHz, 25 °C, $CDCl_3$) of *m*-BTD_2DHPP.

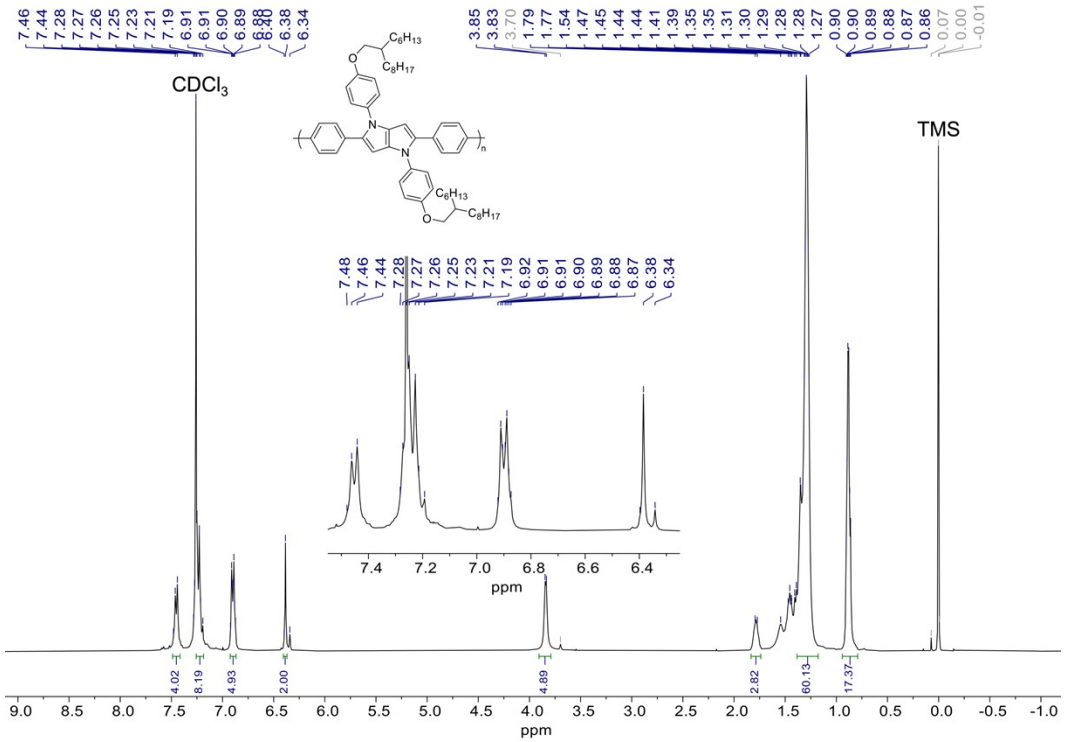


Figure S9. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of Ph_2DHPP.

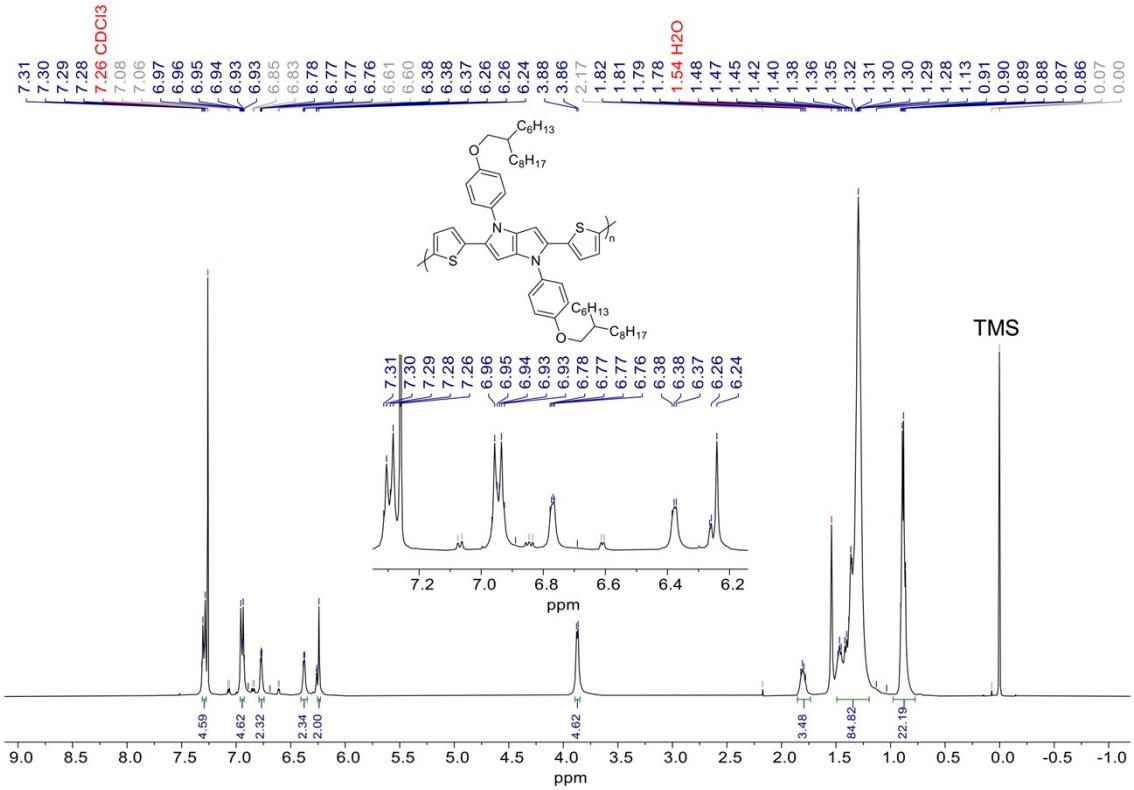


Figure S10. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of Th_2DHPP.

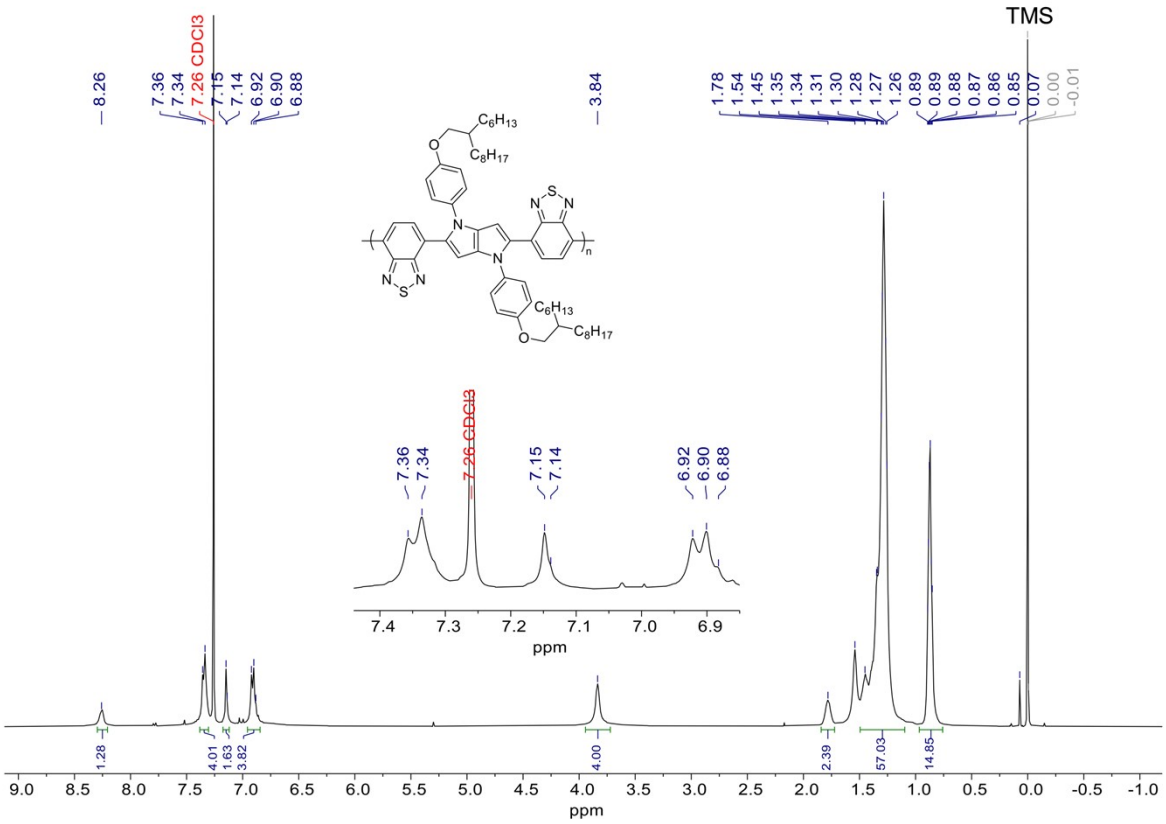


Figure S11. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) of BTD_2DHPP.

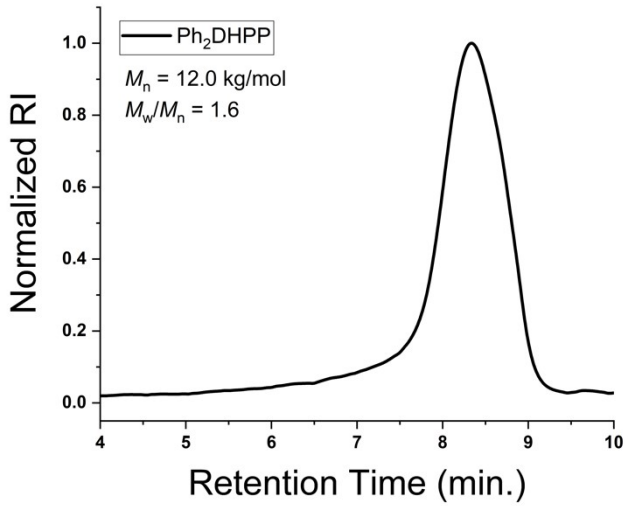


Figure S12. SEC elugram for Ph_2DHPP using $CHCl_3$ as the eluent at 35 °C with a flow rate of 1 mL/min.

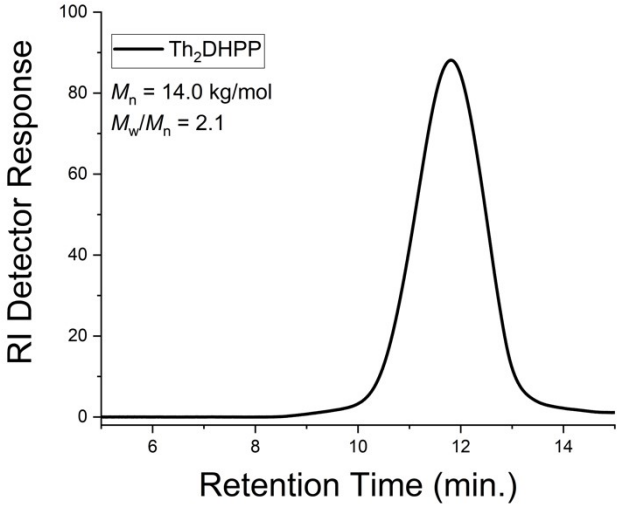


Figure S13. SEC elugram for Th_2DHPP using $CHCl_3$ as the eluent at 35 °C with a flow rate of 1 mL/min.

$$SC = 35\frac{NSS}{NSS_{max}} + 25\frac{log(RY)}{log(RY_{max})} + 15\frac{NUO}{NUO_{max}} + 15\frac{NCC}{NCC_{max}} + 10\frac{NHC}{NHC_{max}} \quad \text{(S1)}$$

The 5 variables in Equation 1 are defined as the number of synthetic steps (NSS), the reciprocal yield of monomers (RY), the number of operations required for purification of monomers (NUO), the number of column chromatography purifications (NCC), and the number of hazardous materials used (NHC), all of which are assigned a weighted value based on the influence each step has on potential cost implications.

Tables S1. Synthetic complexity analysis of DHPP homopolymers.

Polymer	**NSS**	**RY**	**NUO**	**NCC**	**NHC**	**SC**
Ph_2DHPP	4	2.13	3	2	2	14.5
Th_2DHPP	4	6.7	3	2	2	20.9
BTD_2DHPP	4	8.33	4	2	2	22.5

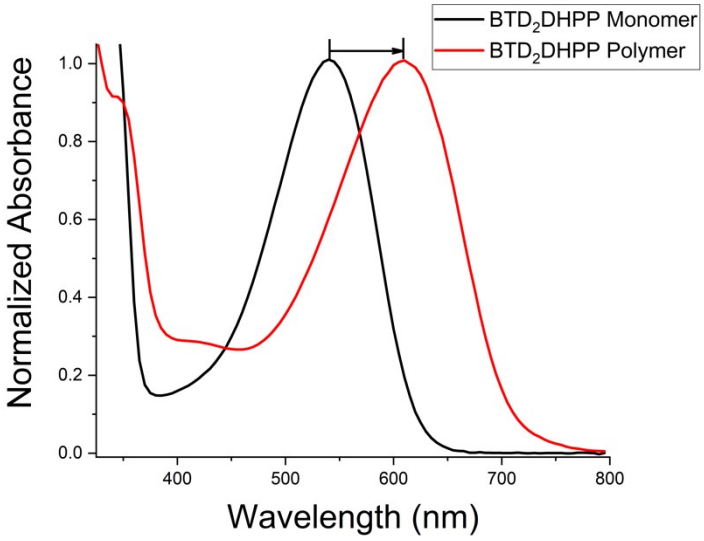


Figure S14. UV-vis absorbance spectra of solutions containing *m*-BTD_2DHPP (black) and BTD_2DHPP (red) to demonstrate the extended effective conjugation length upon polymerization of *m*-BTD_2DHPP.

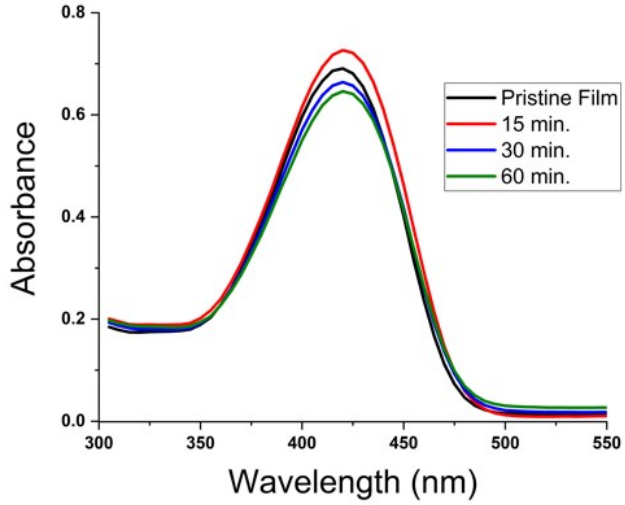


Figure S15. UV-vis absorbance spectra of Ph_2DHPP as a pristine film (black) and after annealing at 100 °C for the time identified in the legend. The lack of change demonstrates the thermal stability of the aggregates in the solid state.

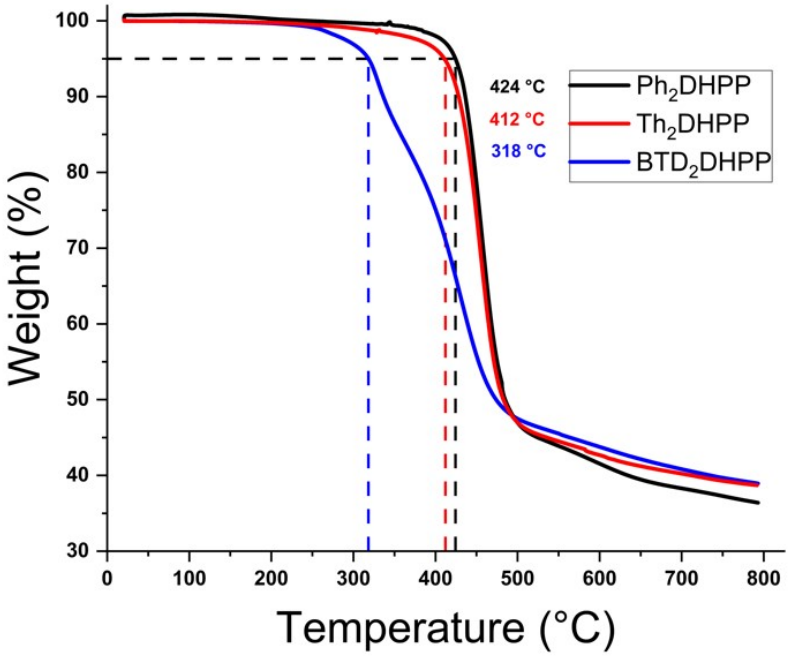


Figure S16. Thermal gravimetric analysis (TGA) traces for Ph2DHPP, Th2DHPP, and BTD2DHPP when heating from 25 °C – 800 °C with a 10 °C/min. heating rate.

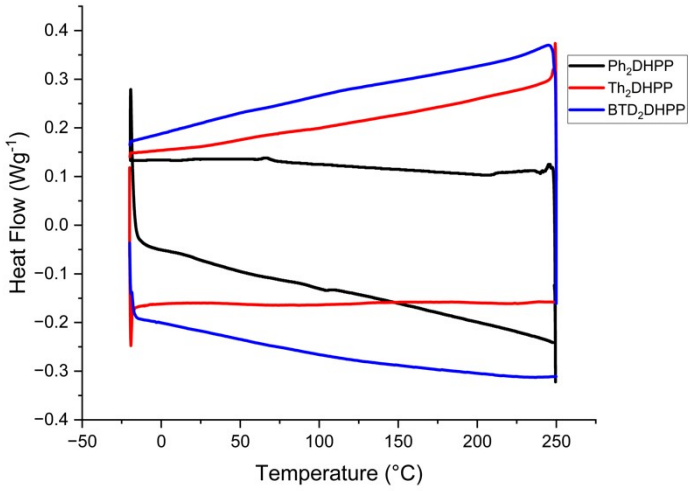


Figure S17. Differential scanning calorimetry (DSC) traces from the second heating cycle for Ph_2DHPP, Th_2DHPP, and BTD_2DHPP.

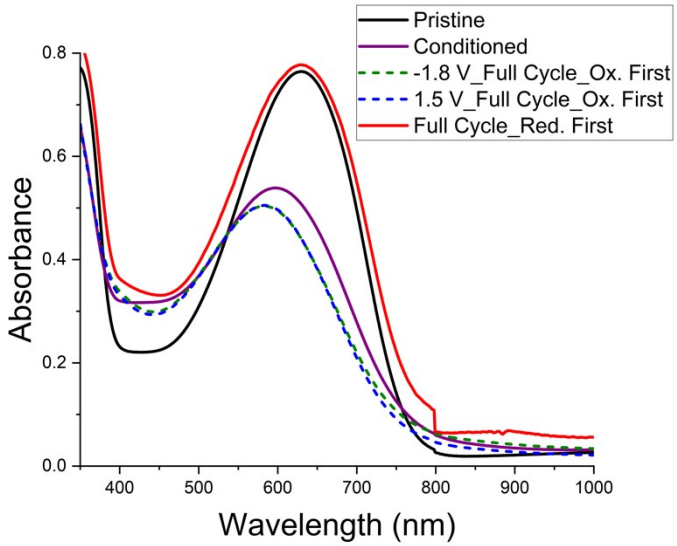


Figure S18. UV-vis absorbance spectra of BTD_2DHPP films after varying exposure to electrochemical processes.

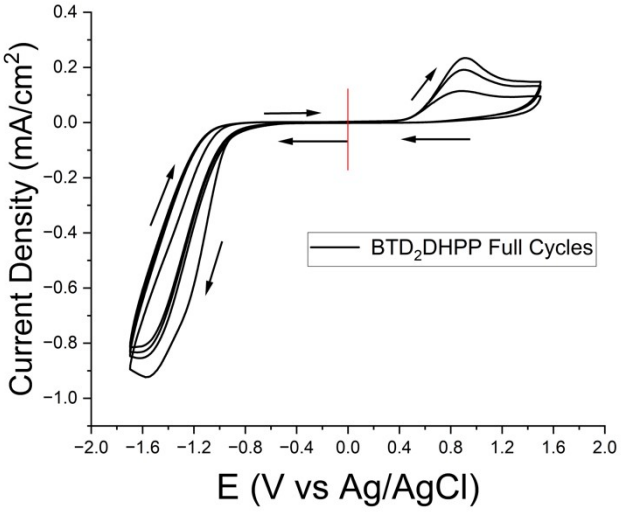


Figure S19. Full CV traces of a BTD_2DHPP sweeping towards reductive potentials first showing the improved electrochemical stability.

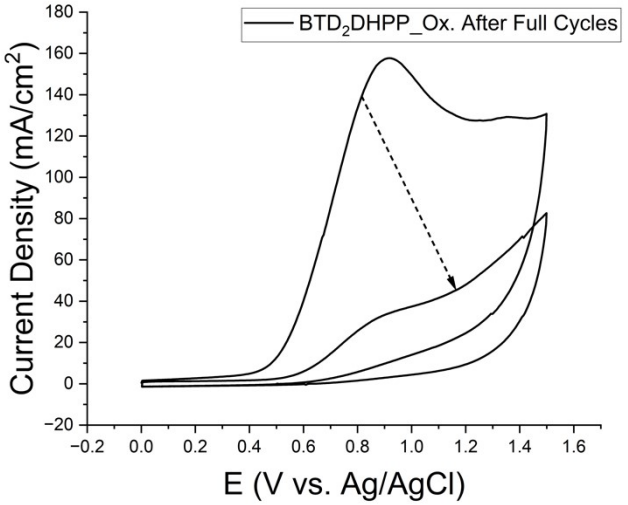


Figure S20. Oxidative CV traces of a BTD_2DHPP film showing the rapid depletion is redox response without the accompanying reductive sweep.

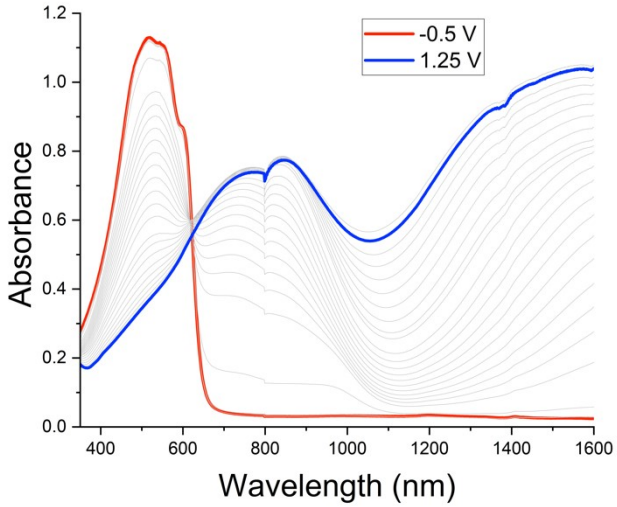


Figure S21. Absorbance as a function of electrochemical potential for a P3HT film by varying potentials in a 0.5 M $TBAPF_6$/ACN electrolyte.

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DFT/TDDFT Gaussian Setups

Optimization Frequency

#opt=(calc,tight) freq geom=connectivity mpw3pbe sv charge = 0 multiplicity = 1 (neutral)
#opt=(calc,tight) freq geom=connectivity mpw3pbe sv charge = 1 multiplicity = 2 (radical+)

TD-SCF

td=(nstates=15) pvdz geom=connectivity mpw3pbe sv charge = 0 multiplicity = 1 (neutral)
td=(nstates=15) pvdz geom=connectivity mpw3pbe sv charge = 1 multiplicity = 2 (radical+)

Ph_2DHPP Neutral

Cartesian Coordinates

C	9.3246	0.8957	0.02715
C	8.42831	1.88399	0.45883
C	7.1262	1.36455	0.23262
C	7.23795	0.08787	-0.33341
C	5.03923	0.5567	-0.12739
C	5.93622	-0.4316	-0.55965
H	8.71324	2.8676	0.82551
H	5.65151	-1.41537	-0.92607
N	5.77356	1.66749	0.35371
N	8.59086	-0.21505	-0.45414
C	5.27243	2.94479	0.73564
C	5.69258	3.52646	1.94752
C	4.3871	3.6497	-0.09177
C	5.23285	4.78932	2.3218
H	6.37791	2.97554	2.59762
C	3.90644	4.9111	0.28763
H	4.07026	3.20978	-1.0405
C	4.33213	5.48608	1.49622
H	5.54909	5.25584	3.25758
H	3.21284	5.43431	-0.37377
C	9.09201	-1.49062	-0.84123
C	9.98409	-2.19558	-0.02119
C	8.66522	-2.07062	-2.05167
C	10.46475	-3.45524	-0.40648
H	10.30627	-1.75715	0.92641
C	9.12491	-3.33176	-2.43161
H	7.97487	-1.51963	-2.69634
C	10.03235	-4.0285	-1.61346
H	11.16388	-3.97844	0.24908
H	8.80363	-3.79672	-3.36644
O	3.92385	6.72392	1.95409
O	10.44008	-5.26462	-2.07669
C	3.0082	7.50696	1.16977
H	2.84218	8.4319	1.74165
H	3.43565	7.75946	0.17999
H	2.04254	6.98301	1.03097

C	11.3645	-6.0462	-1.30149
H	10.94583	-6.30243	-0.30887
H	11.52914	-6.96932	-1.87671
H	12.32946	-5.51924	-1.16921
C	3.57808	0.45548	-0.08285
C	2.80905	0.97662	0.98304
C	2.88889	-0.23483	-1.10651
C	1.42138	0.81905	1.01511
H	3.30639	1.48537	1.81278
C	1.50231	-0.39734	-1.06635
H	3.45339	-0.62462	-1.95903
C	0.73134	0.12971	-0.00661
H	0.86529	1.20776	1.87369
H	1.00363	-0.91081	-1.89405
C	10.78809	0.99687	-0.01681
C	11.47261	1.66901	1.02241
C	11.54841	0.49104	-1.09669
C	12.86251	1.83199	0.98297
H	10.90213	2.05025	1.8749
C	12.93953	0.64681	-1.12813
H	11.04364	-0.00949	-1.92708
C	13.60495	1.31723	-0.09055
H	13.36951	2.35508	1.80006
H	13.5066	0.25083	-1.97658
H	14.69205	1.43913	-0.11953
C	-0.74097	-0.03499	0.03292
C	-1.57667	0.95623	0.59327
C	-1.36591	-1.1895	-0.48788
C	-2.96335	0.79569	0.6402
H	-1.13433	1.87693	0.98588
C	-2.75322	-1.35001	-0.45118
H	-0.75287	-1.98817	-0.91636
C	-3.58731	-0.36126	0.11941
H	-3.58187	1.5746	1.09651
H	-3.19607	-2.25172	-0.88166
C	-5.04874	-0.47145	0.1355
C	-5.97092	0.56342	-0.08144
N	-5.75193	-1.68996	0.29394
C	-7.2562	-0.03914	-0.0577
H	-5.70984	1.59835	-0.29087
C	-7.1095	-1.4149	0.16241
C	-5.22795	-2.95481	0.68686
N	-8.61409	0.23553	-0.18978
C	-8.39503	-2.0177	0.18553

C	-4.39397	-3.07416	1.80734
C	-5.57203	-4.11031	-0.0411
C	-9.31648	-0.98336	-0.03208
C	-9.13887	1.50071	-0.57942
H	-8.65647	-3.05276	0.39395
C	-3.88768	-4.32428	2.19055
H	-4.13659	-2.18302	2.38495
C	-5.08745	-5.35978	0.34655
H	-6.21699	-4.01867	-0.91942
C	-10.77988	-1.09372	-0.01603
C	-8.78829	2.65578	0.14627
C	-9.97918	1.62175	-1.695
C	-4.23683	-5.47255	1.46108
H	-3.23396	-4.38596	3.06293
H	-5.34447	-6.26636	-0.20605
C	-11.39921	-2.24181	-0.56251
C	-11.60511	-0.1119	0.57988
C	-9.27178	3.9063	-0.23925
H	-8.13873	2.56292	1.02106
C	-10.48398	2.87316	-2.07624
H	-10.24167	0.73112	-2.27107
O	-3.79758	-6.74726	1.76144
C	-12.78887	-2.40407	-0.5131
H	-10.77741	-3.00268	-1.04417
C	-12.99551	-0.27244	0.61864
H	-11.15228	0.77326	1.0341
C	-10.12761	4.02086	-1.34951
H	-9.00945	4.81242	0.31163
H	-11.14164	2.93625	-2.94559
C	-2.92221	-6.9533	2.88323
C	-13.59587	-1.41757	0.07358
H	-13.24468	-3.30117	-0.94407
H	-13.61365	0.49875	1.08916
O	-10.56435	5.29702	-1.64877
H	-3.40373	-6.65191	3.83374
H	-2.71666	-8.03374	2.90557
H	-1.97026	-6.40098	2.76114
H	-14.68267	-1.54082	0.10766
C	-11.43989	5.50554	-2.76967
H	-11.64095	6.58682	-2.79313
H	-10.96085	5.20096	-3.72044
H	-12.3942	4.95748	-2.64615

Energy Levels

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Alpha occ. eigenvalues -- -10.20749 -10.20747 -10.20742 -10.20734 -10.19183
Alpha occ. eigenvalues -- -10.19170 -10.19132 -10.19119 -10.19020 -10.19012
Alpha occ. eigenvalues -- -10.18910 -10.18898 -10.18580 -10.18579 -10.18565
Alpha occ. eigenvalues -- -10.18555 -10.18548 -10.18540 -10.18508 -10.18501
Alpha occ. eigenvalues -- -10.18424 -10.18420 -10.18419 -10.18409 -10.18384
Alpha occ. eigenvalues -- -10.18332 -10.18017 -10.18004 -10.17983 -10.17938
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Alpha occ. eigenvalues -- -10.17308 -10.17282 -10.17271 -10.16498 -10.16491
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Alpha occ. eigenvalues -- -0.35498 -0.35442 -0.35270 -0.35002 -0.34708

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Alpha virt. eigenvalues --   0.06186 0.06238 0.07010 0.07096 0.07261
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Alpha virt. eigenvalues --   0.11000 0.11119 0.11121 0.11178 0.11243
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Alpha virt. eigenvalues --   0.12230 0.12286 0.12506 0.12795 0.12854
Alpha virt. eigenvalues --   0.12862 0.12935 0.13021 0.13247 0.13392
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Alpha virt. eigenvalues --   0.16965 0.17049 0.17743 0.18646 0.18814
Alpha virt. eigenvalues --   0.18999 0.19269 0.20118 0.21776 0.21814
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Alpha virt. eigenvalues --   0.23097 0.23301 0.23380 0.24527 0.25322
Alpha virt. eigenvalues --   0.26359 0.26374 0.26747 0.27001 0.27402
Alpha virt. eigenvalues --   0.27484 0.27555 0.27908 0.28079 0.28692
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Alpha virt. eigenvalues --   0.30176 0.30275 0.30422 0.30694 0.30741
Alpha virt. eigenvalues --   0.30949 0.31017 0.31174 0.31543 0.31749
Alpha virt. eigenvalues --   0.32078 0.32135 0.32202 0.32350 0.32434
Alpha virt. eigenvalues --   0.32474 0.32965 0.33768 0.33826 0.33975
Alpha virt. eigenvalues --   0.34488 0.35002 0.35271 0.35427 0.35660
Alpha virt. eigenvalues --   0.36096 0.36210 0.36382 0.36695 0.36996
Alpha virt. eigenvalues --   0.37112 0.37970 0.38112 0.38243 0.38399
Alpha virt. eigenvalues --   0.38716 0.39283 0.39485 0.39977 0.40068
Alpha virt. eigenvalues --   0.40723 0.40777 0.41310 0.41419 0.41725
Alpha virt. eigenvalues --   0.41868 0.42155 0.42554 0.42922 0.43109
Alpha virt. eigenvalues --   0.43849 0.43909 0.43945 0.44157 0.44356
Alpha virt. eigenvalues --   0.44459 0.44509 0.44718 0.44887 0.45098
Alpha virt. eigenvalues --   0.45173 0.45241 0.45422 0.45531 0.45795
Alpha virt. eigenvalues --   0.45947 0.46068 0.46143 0.46332 0.46377

Alpha virt. eigenvalues --	0.46406	0.46678	0.46803	0.47099	0.47393
Alpha virt. eigenvalues --	0.47530	0.47616	0.47701	0.47752	0.48101
Alpha virt. eigenvalues --	0.48164	0.48438	0.48520	0.48682	0.48870
Alpha virt. eigenvalues --	0.49021	0.49486	0.49548	0.49625	0.49733
Alpha virt. eigenvalues --	0.49794	0.49952	0.49985	0.50357	0.50422
Alpha virt. eigenvalues --	0.50492	0.50788	0.50847	0.51299	0.51509
Alpha virt. eigenvalues --	0.52063	0.52095	0.52229	0.52441	0.52596
Alpha virt. eigenvalues --	0.52817	0.52959	0.53267	0.53548	0.53604
Alpha virt. eigenvalues --	0.53968	0.54035	0.54313	0.54703	0.54984
Alpha virt. eigenvalues --	0.55677	0.55800	0.55922	0.56533	0.56648
Alpha virt. eigenvalues --	0.56916	0.57088	0.57392	0.57526	0.57608
Alpha virt. eigenvalues --	0.57793	0.57990	0.58284	0.58433	0.59033
Alpha virt. eigenvalues --	0.59306	0.59439	0.59810	0.60083	0.60306
Alpha virt. eigenvalues --	0.60729	0.61039	0.61267	0.61338	0.61398
Alpha virt. eigenvalues --	0.61710	0.61893	0.62133	0.62448	0.62672
Alpha virt. eigenvalues --	0.63014	0.63167	0.63386	0.63553	0.64026
Alpha virt. eigenvalues --	0.64059	0.64156	0.64271	0.64357	0.64495
Alpha virt. eigenvalues --	0.64686	0.64748	0.65145	0.65505	0.65814
Alpha virt. eigenvalues --	0.65822	0.65861	0.65947	0.66288	0.66357
Alpha virt. eigenvalues --	0.66686	0.66729	0.66742	0.66943	0.66990
Alpha virt. eigenvalues --	0.67158	0.67393	0.67450	0.67575	0.68079
Alpha virt. eigenvalues --	0.68160	0.68209	0.68271	0.68340	0.68413
Alpha virt. eigenvalues --	0.68617	0.68857	0.69163	0.69267	0.69378
Alpha virt. eigenvalues --	0.69433	0.69530	0.69645	0.69681	0.69729
Alpha virt. eigenvalues --	0.69911	0.69989	0.70188	0.70383	0.70508
Alpha virt. eigenvalues --	0.70606	0.70892	0.70933	0.71001	0.71196
Alpha virt. eigenvalues --	0.71294	0.71669	0.71997	0.72218	0.72357
Alpha virt. eigenvalues --	0.72609	0.72838	0.72969	0.73053	0.73258
Alpha virt. eigenvalues --	0.73506	0.73638	0.73852	0.74118	0.74543
Alpha virt. eigenvalues --	0.74836	0.74974	0.75095	0.75248	0.75500
Alpha virt. eigenvalues --	0.75508	0.75720	0.75800	0.76053	0.76483
Alpha virt. eigenvalues --	0.77014	0.77444	0.77592	0.77843	0.78003
Alpha virt. eigenvalues --	0.78085	0.78265	0.78444	0.78577	0.78862
Alpha virt. eigenvalues --	0.79136	0.79213	0.79443	0.79837	0.80231
Alpha virt. eigenvalues --	0.80530	0.80774	0.80824	0.80983	0.81217
Alpha virt. eigenvalues --	0.81301	0.81811	0.82655	0.82820	0.82931
Alpha virt. eigenvalues --	0.83213	0.83269	0.84098	0.84249	0.84417
Alpha virt. eigenvalues --	0.84756	0.84992	0.85146	0.85342	0.85657
Alpha virt. eigenvalues --	0.85760	0.85910	0.86019	0.87374	0.88327
Alpha virt. eigenvalues --	0.88573	0.88626	0.89342	0.89622	0.89693
Alpha virt. eigenvalues --	0.89887	0.91219	0.91527	0.91810	0.92096
Alpha virt. eigenvalues --	0.92358	0.93454	0.93474	0.93800	0.94186
Alpha virt. eigenvalues --	0.94205	0.95095	0.95269	0.95585	0.96099
Alpha virt. eigenvalues --	0.97078	0.97334	0.98206	0.98882	0.99160

Alpha virt. eigenvalues --	0.99369	0.99611	1.00154	1.00500	1.01358
Alpha virt. eigenvalues --	1.01769	1.01809	1.02262	1.02357	1.02974
Alpha virt. eigenvalues --	1.03754	1.04026	1.05450	1.06684	1.07049
Alpha virt. eigenvalues --	1.07591	1.08159	1.10103	1.10219	1.10568
Alpha virt. eigenvalues --	1.10591	1.10975	1.11141	1.12137	1.12539
Alpha virt. eigenvalues --	1.12766	1.13072	1.13517	1.13837	1.13946
Alpha virt. eigenvalues --	1.14466	1.14624	1.15168	1.15486	1.15949
Alpha virt. eigenvalues --	1.16074	1.16568	1.16710	1.17080	1.17167
Alpha virt. eigenvalues --	1.17491	1.17541	1.18038	1.18759	1.19807
Alpha virt. eigenvalues --	1.21533	1.22694	1.22812	1.23100	1.24824
Alpha virt. eigenvalues --	1.25046	1.25338	1.27375	1.27558	1.27611
Alpha virt. eigenvalues --	1.27809	1.27925	1.28872	1.30565	1.30744
Alpha virt. eigenvalues --	1.32048	1.33188	1.33976	1.33995	1.34858
Alpha virt. eigenvalues --	1.35805	1.36679	1.37467	1.37868	1.38576
Alpha virt. eigenvalues --	1.41035	1.44608	1.47099	1.48575	1.49996
Alpha virt. eigenvalues --	1.50157	1.50967	1.52362	1.52447	1.53586
Alpha virt. eigenvalues --	1.57649	1.57774	1.58445	1.58527	1.58586
Alpha virt. eigenvalues --	1.58823	1.58975	1.59890	1.65666	1.67038
Alpha virt. eigenvalues --	1.73960	1.73976	1.74008	1.74060	1.77044
Alpha virt. eigenvalues --	1.77581				

Lowest Excited States (ES)

Table S2. A table of the lowest lying 15 excited states where the energy (eV), oscillator strengths (f), most significant transition, % contribution of the transition are provided for each.

ES	Energy (eV)	*f*	Transition	% Contribution
1	3.00	2.1848	HOMO → LUMO	100%
2	3.33	0.0012	HOMO – 1 → LUMO	87.2%
3	3.61	0.0042	HOMO → LUMO + 1	63.0%
4	3.68	0.1178	HOMO – 2 → LUMO	86.0%
5	3.70	0.0080	HOMO – 3 → LUMO	67.5%
6	3.70	0.0689	HOMO → LUMO + 2	42.1%
7	3.71	0.0007	HOMO → LUMO + 3	36.8%
8	3.72	0.0040	HOMO → LUMO + 4	36.7%
9	3.73	0.0047	HOMO → LUMO + 5	40.8%
10	3.83	0.5231	HOMO – 1 → LUMO+1	56.9%
11	3.90	0.0303	HOMO → LUMO + 6	28.8%
12	3.98	0.0001	HOMO → LUMO + 7	58.3%
13	4.03	0.0068	HOMO → LUMO + 8	44.3%
14	4.03	0.0002	HOMO – 1 → LUMO+2	32.4%
15	4.09	0.0718	HOMO → LUMO + 10	44.2%

Geometric Images

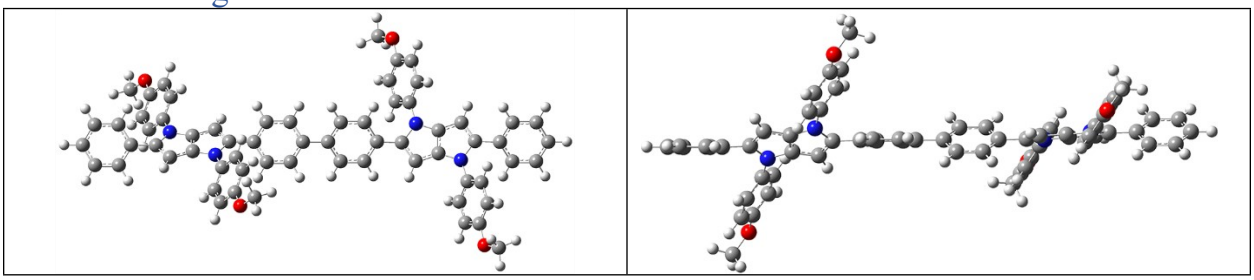

Figure S22. Two geometric perspectives are given with a top view on the left and a front view on the right.

Simulated UV-Vis Spectra

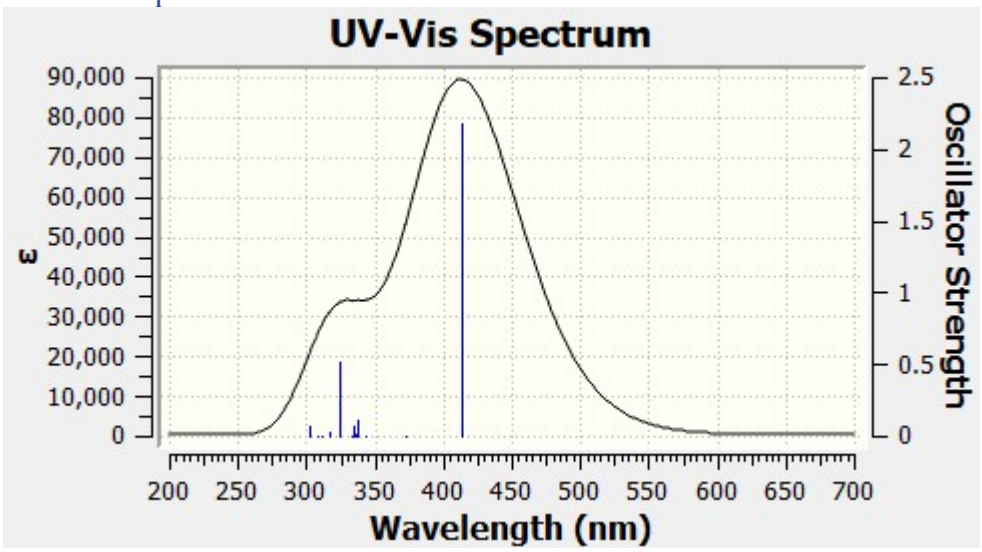

Figure S23. Simulated UV-visible absorbance spectrum of the neutral Ph_2DHPP oligomer in gas phase.

Ph_2DHPP Radical Cation

Cartesian Coordinates

C	9.36346	0.87422	0.0496
C	8.45935	1.89549	0.44146
C	7.16368	1.38848	0.22396
C	7.25182	0.06395	-0.29943
C	5.04799	0.58431	-0.12823
C	5.96041	-0.43867	-0.52297
H	8.75318	2.887	0.79004
H	5.67005	-1.43144	-0.87193
N	5.79155	1.71708	0.33803
N	8.62865	-0.2614	-0.40939
C	5.31035	3.07551	0.60138
C	5.71542	3.73411	1.78519

C	4.50141	3.74596	-0.33694
C	5.30611	5.04435	2.02533
H	6.34992	3.20867	2.50965
C	4.07673	5.05881	-0.09704
H	4.20294	3.23489	-1.2608
C	4.47768	5.72706	1.08898
H	5.61138	5.57043	2.93784
H	3.44836	5.55899	-0.83923
C	9.13038	-1.60302	-0.71004
C	10.02618	-2.24831	0.16486
C	8.67242	-2.26805	-1.87082
C	10.48462	-3.54122	-0.11716
H	10.36818	-1.73318	1.07103
C	9.11421	-3.55949	-2.15081
H	7.97496	-1.76089	-2.54862
C	10.03162	-4.21551	-1.2807
H	11.18314	-4.02038	0.57452
H	8.77087	-4.08897	-3.04761
O	4.13963	7.0381	1.46391
O	10.39516	-5.50885	-1.69146
C	3.3062	7.72321	0.46801
H	3.12459	8.73144	0.88136
H	3.82054	7.82038	-0.50868
H	2.33361	7.21365	0.31809
C	11.34305	-6.15648	-0.7759
H	10.91637	-6.2904	0.23801
H	11.54411	-7.14949	-1.21616
H	12.29571	-5.59577	-0.69879
C	3.58352	0.47228	-0.08641
C	2.77535	1.23353	0.80615
C	2.92777	-0.47655	-0.92422
C	1.39368	1.06214	0.84505
H	3.24947	1.94462	1.49168
C	1.54669	-0.64435	-0.87954
H	3.51999	-1.06834	-1.63218
C	0.73523	0.12183	0.00387
H	0.80954	1.64533	1.56633
H	1.07796	-1.36401	-1.56057
C	10.84446	0.97281	0.02187
C	11.50208	1.77993	0.98551
C	11.622	0.33575	-0.97892
C	12.8942	1.9302	0.95807
H	10.91268	2.27732	1.76537
C	13.01355	0.49425	-1.0025

H	11.12853	-0.26873	-1.74876
C	13.65501	1.28691	-0.03413
H	13.39015	2.55097	1.71494
H	13.60259	0.00204	-1.78669
H	14.74583	1.40692	-0.05564
C	-0.74325	-0.05695	0.0506
C	-1.59766	0.97241	0.53624
C	-1.35798	-1.26112	-0.39288
C	-2.97975	0.80963	0.57336
H	-1.16624	1.923	0.87045
C	-2.73897	-1.43654	-0.34239
H	-0.73535	-2.0802	-0.77104
C	-3.5911	-0.40459	0.14495
H	-3.60903	1.62442	0.95066
H	-3.17619	-2.3756	-0.69941
C	-5.056	-0.52246	0.16247
C	-5.98874	0.5535	0.07724
N	-5.77323	-1.7627	0.15237
C	-7.26655	-0.02411	0.0148
H	-5.71785	1.60913	0.01464
C	-7.15015	-1.44534	0.06543
C	-5.27624	-3.10361	0.47026
N	-8.64773	0.28866	-0.07538
C	-8.43244	-2.02461	0.00951
C	-4.50801	-3.32953	1.6292
C	-5.62482	-4.18618	-0.36987
C	-9.35683	-0.95121	-0.07783
C	-9.166	1.62821	-0.35678
H	-8.70681	-3.0791	0.07072
C	-4.06756	-4.62034	1.94734
H	-4.25337	-2.48777	2.28523
C	-5.19995	-5.47507	-0.05346
H	-6.22774	-4.00484	-1.26827
C	-10.83718	-1.06213	-0.07989
C	-8.77012	2.70904	0.46444
C	-10.01547	1.8536	-1.45766
C	-4.41171	-5.71255	1.10908
H	-3.47096	-4.7715	2.85127
H	-5.46148	-6.32624	-0.69357
C	-11.44491	-2.19962	-0.67094
C	-11.66447	-0.09614	0.54818
C	-9.22725	3.99562	0.18664
H	-8.10859	2.52777	1.32036
C	-10.48921	3.14198	-1.73508

H	-10.30902	1.01379	-2.09955
O	-4.05157	-7.05702	1.29911
C	-12.83646	-2.35588	-0.64873
H	-10.81675	-2.95359	-1.16075
C	-13.05524	-0.26148	0.57018
H	-11.2108	0.7749	1.035
C	-10.09833	4.23236	-0.9153
H	-8.93191	4.84487	0.81434
H	-11.15096	3.29206	-2.59271
C	-3.2586	-7.27257	2.51594
C	-13.64661	-1.38722	-0.03024
H	-13.29327	-3.23676	-1.11728
H	-13.68326	0.48948	1.06601
O	-10.48394	5.5743	-1.07045
H	-3.81462	-6.98629	3.43071
H	-3.05358	-8.35772	2.54383
H	-2.29636	-6.72363	2.48798
H	-14.73698	-1.51177	-0.01195
C	-11.38274	5.78466	-2.21244
H	-11.60713	6.86632	-2.21367
H	-10.9034	5.51393	-3.17417
H	-12.32974	5.21925	-2.10579

Energy Levels

Alpha occ. eigenvalues -- -19.22802 -19.22761 -19.22342 -19.22318 -14.46353
Alpha occ. eigenvalues -- -14.46333 -14.46140 -14.46120 -10.31668 -10.31637
Alpha occ. eigenvalues -- -10.31611 -10.31571 -10.31349 -10.31325 -10.31194
Alpha occ. eigenvalues -- -10.31169 -10.30197 -10.30182 -10.29804 -10.29783
Alpha occ. eigenvalues -- -10.29751 -10.29730 -10.29578 -10.29556 -10.28812
Alpha occ. eigenvalues -- -10.28754 -10.28484 -10.28472 -10.28327 -10.28308
Alpha occ. eigenvalues -- -10.27890 -10.27881 -10.26974 -10.26957 -10.26663
Alpha occ. eigenvalues -- -10.26640 -10.26513 -10.26482 -10.26277 -10.26272
Alpha occ. eigenvalues -- -10.26259 -10.26241 -10.26234 -10.26220 -10.26204
Alpha occ. eigenvalues -- -10.26200 -10.26036 -10.26019 -10.25862 -10.25838
Alpha occ. eigenvalues -- -10.25793 -10.25760 -10.25739 -10.25717 -10.25665
Alpha occ. eigenvalues -- -10.25638 -10.25290 -10.25269 -10.25186 -10.25161
Alpha occ. eigenvalues -- -10.25080 -10.25065 -10.24946 -10.24929 -10.24785
Alpha occ. eigenvalues -- -10.24757 -10.24596 -10.24580 -10.24491 -10.24477
Alpha occ. eigenvalues -- -10.24444 -10.24429 -1.12033 -1.11992 -1.11568
Alpha occ. eigenvalues -- -1.11545 -1.08181 -1.08159 -1.05011 -1.04990
Alpha occ. eigenvalues -- -0.95749 -0.94312 -0.94244 -0.94092 -0.93547
Alpha occ. eigenvalues -- -0.93452 -0.92915 -0.92877 -0.89152 -0.88458
Alpha occ. eigenvalues -- -0.88245 -0.88114 -0.87175 -0.85028 -0.83865
Alpha occ. eigenvalues -- -0.83773 -0.83302 -0.83277 -0.82893 -0.82881

```
Alpha  occ. eigenvalues --   -0.82775  -0.82554  -0.81633  -0.81621  -0.81611
Alpha  occ. eigenvalues --   -0.81402  -0.80623  -0.80538  -0.78521  -0.76743
Alpha  occ. eigenvalues --   -0.76582  -0.76541  -0.75816  -0.75807  -0.73350
Alpha  occ. eigenvalues --   -0.71576  -0.70965  -0.70630  -0.70331  -0.69827
Alpha  occ. eigenvalues --   -0.69434  -0.69365  -0.69102  -0.68618  -0.68299
Alpha  occ. eigenvalues --   -0.68215  -0.67903  -0.67789  -0.67532  -0.66746
Alpha  occ. eigenvalues --   -0.65179  -0.65160  -0.64721  -0.64717  -0.63668
Alpha  occ. eigenvalues --   -0.63608  -0.62730  -0.62624  -0.61242  -0.60308
Alpha  occ. eigenvalues --   -0.60259  -0.59714  -0.59341  -0.59316  -0.58587
Alpha  occ. eigenvalues --   -0.57830  -0.57685  -0.57275  -0.56914  -0.56429
Alpha  occ. eigenvalues --   -0.56166  -0.55933  -0.55805  -0.55656  -0.54594
Alpha  occ. eigenvalues --   -0.54238  -0.54218  -0.53424  -0.53253  -0.53178
Alpha  occ. eigenvalues --   -0.53129  -0.52864  -0.52846  -0.52831  -0.52709
Alpha  occ. eigenvalues --   -0.52665  -0.52490  -0.52457  -0.52443  -0.52420
Alpha  occ. eigenvalues --   -0.52233  -0.52095  -0.51706  -0.51397  -0.51365
Alpha  occ. eigenvalues --   -0.51088  -0.50980  -0.50940  -0.50583  -0.50540
Alpha  occ. eigenvalues --   -0.50475  -0.49756  -0.49691  -0.49643  -0.49200
Alpha  occ. eigenvalues --   -0.48964  -0.48921  -0.48435  -0.48336  -0.48170
Alpha  occ. eigenvalues --   -0.48146  -0.47998  -0.47503  -0.47302  -0.47105
Alpha  occ. eigenvalues --   -0.46951  -0.46601  -0.46326  -0.46062  -0.46018
Alpha  occ. eigenvalues --   -0.45796  -0.45698  -0.45470  -0.44880  -0.44247
Alpha  occ. eigenvalues --   -0.44201  -0.44092  -0.44041  -0.43932  -0.43806
Alpha  occ. eigenvalues --   -0.43130  -0.42872  -0.42671  -0.42644  -0.41932
Alpha  occ. eigenvalues --   -0.41806  -0.41792  -0.40572  -0.40430  -0.40271
Alpha  occ. eigenvalues --   -0.40017  -0.39995  -0.39799  -0.39620  -0.39420
Alpha  occ. eigenvalues --   -0.38209  -0.37316  -0.36945  -0.35186  -0.35071
Alpha  occ. eigenvalues --   -0.34839  -0.34176  -0.34112  -0.34080  -0.33995
Alpha  occ. eigenvalues --   -0.33427  -0.32942  -0.32923  -0.32631  -0.32615
Alpha  occ. eigenvalues --   -0.31832  -0.29918  -0.29819  -0.28672  -0.28563
Alpha  occ. eigenvalues --   -0.27628  -0.26565
Alpha virt. eigenvalues --   -0.15038  -0.12586  -0.10568  -0.10369  -0.09538
Alpha virt. eigenvalues --   -0.09263  -0.08993  -0.08957  -0.08715  -0.08675
Alpha virt. eigenvalues --   -0.08153  -0.07960  -0.07922  -0.07759  -0.07453
Alpha virt. eigenvalues --   -0.07435  -0.04866  -0.03133  -0.02149  -0.01555
Alpha virt. eigenvalues --   -0.00643  -0.00346  -0.00012   0.00023   0.00326
Alpha virt. eigenvalues --    0.00392   0.00496   0.00681   0.01228   0.01286
Alpha virt. eigenvalues --    0.01557   0.01614   0.01666   0.01732   0.02168
Alpha virt. eigenvalues --    0.02409   0.02761   0.03125   0.03679   0.03718
Alpha virt. eigenvalues --    0.03814   0.03920   0.03976   0.04118   0.04334
Alpha virt. eigenvalues --    0.04367   0.04613   0.05005   0.05139   0.05172
Alpha virt. eigenvalues --    0.05223   0.05907   0.06100   0.06442   0.06459
Alpha virt. eigenvalues --    0.06532   0.06665   0.06690   0.06765   0.07000
Alpha virt. eigenvalues --    0.07009   0.07101   0.07127   0.07200   0.07263
Alpha virt. eigenvalues --    0.07420   0.07467   0.07738   0.07752   0.08020
```

Alpha virt. eigenvalues --	0.08113	0.08229	0.08271	0.08823	0.09109
Alpha virt. eigenvalues --	0.09243	0.09574	0.09618	0.09786	0.09833
Alpha virt. eigenvalues --	0.10229	0.10297	0.10392	0.10405	0.10656
Alpha virt. eigenvalues --	0.10756	0.11560	0.12187	0.12693	0.12905
Alpha virt. eigenvalues --	0.13443	0.13842	0.13907	0.14198	0.14309
Alpha virt. eigenvalues --	0.14731	0.14755	0.15116	0.15659	0.16063
Alpha virt. eigenvalues --	0.17208	0.17532	0.18247	0.18629	0.18684
Alpha virt. eigenvalues --	0.19544	0.19647	0.19837	0.20076	0.20248
Alpha virt. eigenvalues --	0.20654	0.21369	0.21403	0.22002	0.22172
Alpha virt. eigenvalues --	0.22464	0.22697	0.22792	0.23070	0.23328
Alpha virt. eigenvalues --	0.23406	0.23811	0.23854	0.23881	0.24152
Alpha virt. eigenvalues --	0.24187	0.24258	0.24306	0.24538	0.24764
Alpha virt. eigenvalues --	0.24814	0.25209	0.25480	0.26091	0.26246
Alpha virt. eigenvalues --	0.26506	0.26748	0.27099	0.27208	0.27532
Alpha virt. eigenvalues --	0.27701	0.28396	0.28454	0.28799	0.28823
Alpha virt. eigenvalues --	0.29652	0.29975	0.30224	0.30327	0.30623
Alpha virt. eigenvalues --	0.30998	0.31272	0.31358	0.31977	0.32167
Alpha virt. eigenvalues --	0.32343	0.32523	0.33097	0.33241	0.33860
Alpha virt. eigenvalues --	0.34045	0.34197	0.34360	0.35165	0.35226
Alpha virt. eigenvalues --	0.35537	0.35647	0.35992	0.36270	0.36532
Alpha virt. eigenvalues --	0.36885	0.36985	0.37048	0.37098	0.37236
Alpha virt. eigenvalues --	0.37667	0.37904	0.38205	0.38297	0.38394
Alpha virt. eigenvalues --	0.38462	0.38733	0.39061	0.39195	0.39462
Alpha virt. eigenvalues --	0.39542	0.39857	0.40012	0.40223	0.40331
Alpha virt. eigenvalues --	0.40425	0.40468	0.40629	0.40782	0.40854
Alpha virt. eigenvalues --	0.40925	0.41135	0.41152	0.41287	0.41548
Alpha virt. eigenvalues --	0.41683	0.41737	0.41954	0.41962	0.42160
Alpha virt. eigenvalues --	0.42271	0.42597	0.42771	0.42784	0.42935
Alpha virt. eigenvalues --	0.43053	0.43338	0.43514	0.43540	0.43768
Alpha virt. eigenvalues --	0.43982	0.44312	0.44497	0.44527	0.44596
Alpha virt. eigenvalues --	0.45023	0.45092	0.45222	0.45507	0.45572
Alpha virt. eigenvalues --	0.45906	0.46354	0.46499	0.46516	0.46647
Alpha virt. eigenvalues --	0.46806	0.47491	0.47839	0.47866	0.48394
Alpha virt. eigenvalues --	0.48780	0.48849	0.49245	0.49726	0.49889
Alpha virt. eigenvalues --	0.50269	0.50398	0.50831	0.50861	0.51025
Alpha virt. eigenvalues --	0.51185	0.51440	0.51940	0.51999	0.52133
Alpha virt. eigenvalues --	0.52230	0.52667	0.53195	0.53277	0.53404
Alpha virt. eigenvalues --	0.53633	0.54212	0.54278	0.54376	0.54609
Alpha virt. eigenvalues --	0.54926	0.55120	0.55752	0.55966	0.56194
Alpha virt. eigenvalues --	0.56314	0.56881	0.57043	0.57362	0.57528
Alpha virt. eigenvalues --	0.58010	0.58266	0.58523	0.58571	0.58716
Alpha virt. eigenvalues --	0.58730	0.58955	0.59053	0.59066	0.59255
Alpha virt. eigenvalues --	0.59503	0.59695	0.59895	0.59940	0.60177
Alpha virt. eigenvalues --	0.60314	0.60408	0.60494	0.60820	0.60976

Alpha virt. eigenvalues -- 0.61096 0.61156 0.61217 0.61261 0.61334
Alpha virt. eigenvalues -- 0.61422 0.61530 0.61629 0.61705 0.62036
Alpha virt. eigenvalues -- 0.62200 0.62266 0.62587 0.62655 0.62958
Alpha virt. eigenvalues -- 0.63014 0.63274 0.63345 0.63361 0.63437
Alpha virt. eigenvalues -- 0.63500 0.63659 0.63863 0.63942 0.64006
Alpha virt. eigenvalues -- 0.64114 0.64173 0.64387 0.64483 0.64759
Alpha virt. eigenvalues -- 0.64860 0.65054 0.65269 0.65465 0.65629
Alpha virt. eigenvalues -- 0.65872 0.66114 0.66334 0.66434 0.66883
Alpha virt. eigenvalues -- 0.67033 0.67257 0.67286 0.67399 0.67562
Alpha virt. eigenvalues -- 0.67844 0.68024 0.68268 0.68357 0.68555
Alpha virt. eigenvalues -- 0.68931 0.69743 0.69827 0.70005 0.70152
Alpha virt. eigenvalues -- 0.70425 0.70506 0.70581 0.70862 0.71386
Alpha virt. eigenvalues -- 0.71506 0.71709 0.71845 0.72170 0.72393
Alpha virt. eigenvalues -- 0.72647 0.72681 0.72874 0.73107 0.73152
Alpha virt. eigenvalues -- 0.73770 0.73786 0.74124 0.74360 0.75401
Alpha virt. eigenvalues -- 0.75479 0.75510 0.75836 0.76524 0.76634
Alpha virt. eigenvalues -- 0.77047 0.77149 0.77304 0.77331 0.77396
Alpha virt. eigenvalues -- 0.77730 0.77767 0.78316 0.78679 0.80214
Alpha virt. eigenvalues -- 0.80528 0.80607 0.80898 0.81070 0.81212
Alpha virt. eigenvalues -- 0.81542 0.82299 0.83504 0.83798 0.84075
Alpha virt. eigenvalues -- 0.84315 0.84778 0.85810 0.85931 0.86438
Alpha virt. eigenvalues -- 0.86781 0.86845 0.87488 0.88390 0.88579
Alpha virt. eigenvalues -- 0.89900 0.90536 0.91122 0.91366 0.91753
Alpha virt. eigenvalues -- 0.92080 0.92390 0.92593 0.93703 0.95089
Alpha virt. eigenvalues -- 0.95473 0.95765 0.95889 0.96372 0.96554
Alpha virt. eigenvalues -- 0.97083 0.97985 0.98647 0.99038 1.00448
Alpha virt. eigenvalues -- 1.02576 1.02861 1.03385 1.03444 1.03818
Alpha virt. eigenvalues -- 1.03868 1.04017 1.04277 1.04325 1.04382
Alpha virt. eigenvalues -- 1.05045 1.05204 1.05866 1.06031 1.06662
Alpha virt. eigenvalues -- 1.06801 1.07746 1.07949 1.08560 1.08694
Alpha virt. eigenvalues -- 1.08960 1.09338 1.09814 1.10184 1.10767
Alpha virt. eigenvalues -- 1.10866 1.11344 1.11754 1.11995 1.12998
Alpha virt. eigenvalues -- 1.13413 1.14111 1.14903 1.15894 1.16859
Alpha virt. eigenvalues -- 1.17094 1.17903 1.18221 1.19057 1.19718
Alpha virt. eigenvalues -- 1.19953 1.20165 1.20230 1.23207 1.23550
Alpha virt. eigenvalues -- 1.23792 1.25521 1.25781 1.26968 1.27074
Alpha virt. eigenvalues -- 1.27410 1.28690 1.28969 1.29934 1.30167
Alpha virt. eigenvalues -- 1.32017 1.35720 1.38088 1.40358 1.41842
Alpha virt. eigenvalues -- 1.42150 1.42847 1.43988 1.44826 1.45151
Alpha virt. eigenvalues -- 1.46910 1.47132 1.47705 1.47749 1.50035
Alpha virt. eigenvalues -- 1.50674 1.51063 1.51152 1.56040 1.57344
Alpha virt. eigenvalues -- 1.67353 1.67682 1.68477 1.68544 1.68961
Alpha virt. eigenvalues -- 1.68986

Lowest Excited States (ES)

Table S3. A table of the lowest lying 15 excited states where the energy (eV), oscillator strengths (f), most significant transition, % contribution of the transition are provided for each.

ES	Energy (eV)	*f*	Transition	% Contribution
1	0.67	0.8681	$SOMO_\beta \rightarrow LUMO_\beta$	68.8%
2	0.92	0.0003	$SOMO_\beta - 1 \rightarrow LUMO_\beta$	84.5%
3	0.93	0.0311	$SOMO_\beta - 2 \rightarrow LUMO_\beta$	84.9%
4	1.36	0.0001	$SOMO_\beta - 3 \rightarrow LUMO_\beta$	100%
5	1.38	0.0090	$SOMO_\beta - 2 \rightarrow LUMO_\beta$ $SOMO_\beta - 4 \rightarrow LUMO_\beta$	50.0% 50.0%
6	1.76	0.0001	$SOMO_\beta - 5 \rightarrow LUMO_\beta$	79.6%
7	2.00	0.0001	$SOMO_\beta - 6 \rightarrow LUMO_\beta$	63.6%
8	2.01	0.0000	$SOMO_\beta - 7 \rightarrow LUMO_\beta$	63.5%
9	2.18	0.0322	$SOMO_\beta - 8 \rightarrow LUMO_\beta$	64.0%
10	2.20	0.0002	$SOMO_\beta - 9 \rightarrow LUMO_\beta$	83.9%
11	2.21	0.0057	$SOMO_\beta - 10 \rightarrow LUMO_\beta$	59.5%
12	2.23	0.0003	$SOMO_\beta \rightarrow LUMO_\beta + 1$	53.2%
13	2.35	1.0560	$SOMO_\alpha \rightarrow LUMO_\alpha$	45.6%
14	2.39	0.0010	$SOMO_\beta - 12 \rightarrow LUMO_\beta$	51.7%
15	2.46	0.0000	$SOMO_\beta - 14 \rightarrow LUMO_\beta$	46.9%

Geometric Images

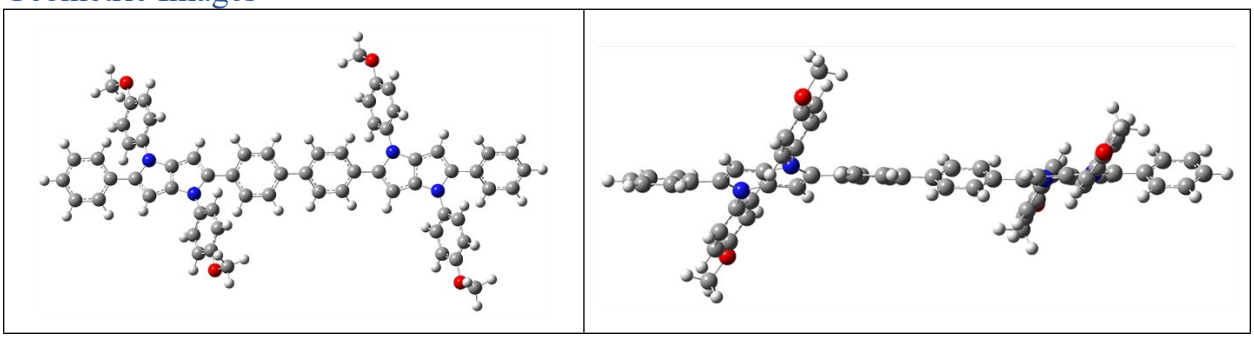

Figure S24. Two geometric perspectives are given with a top view on the left and a front view on the right.

Simulated UV-Vis Spectra

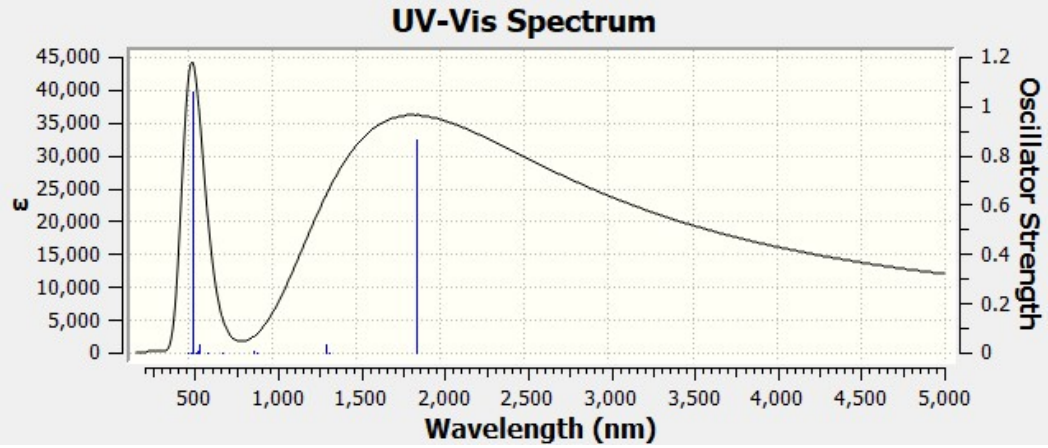

Figure S25. Simulated UV-visible absorbance spectrum of the neutral Ph_2DHPP+1 oligomer in gas phase.

Th_2DHPP Neutral

Cartesian Coordinates

C	8.88335	-0.68491	0.11504
C	8.10267	-1.84712	-0.01028
C	6.7552	-1.41361	-0.03565
C	6.72158	-0.01413	0.07886
C	4.59271	-0.74418	-0.07242
C	5.37479	0.41924	0.05362
H	8.49315	-2.86233	-0.02762
H	4.98451	1.43462	0.06936
N	5.44724	-1.87318	-0.11554
N	8.0304	0.44455	0.15809
C	5.07605	-3.2499	-0.10302
C	5.53255	-4.10807	-1.12122
C	4.28423	-3.76885	0.93059
C	5.19865	-5.46277	-1.10388
H	6.14732	-3.7022	-1.92942
C	3.92999	-5.12512	0.94468
H	3.93857	-3.10619	1.72797
C	4.39036	-5.9768	-0.07367
H	5.54462	-6.14472	-1.88388
H	3.30634	-5.50207	1.75777
C	8.40283	1.8205	0.15133
C	9.2174	2.33819	-0.86506
C	7.92418	2.68038	1.15801
C	9.57122	3.6947	-0.87355
H	9.58001	1.67469	-1.65407

C	8.25787	4.03515	1.14583
H	7.29187	2.27585	1.95325
C	9.08813	4.54794	0.13276
H	10.21195	4.07073	-1.67372
H	7.89451	4.71809	1.91703
O	4.10342	-7.325	-0.14571
O	9.37245	5.8969	0.20852
C	3.28448	-7.93619	0.86628
H	3.20404	-8.99605	0.58312
H	3.75183	-7.8582	1.86699
H	2.27327	-7.48606	0.89515
C	10.20951	6.50722	-0.78869
H	9.76202	6.42585	-1.79821
H	10.28246	7.56798	-0.50676
H	11.22211	6.05921	-0.79703
C	3.16014	-0.82599	-0.20162
C	2.35783	-1.75128	-0.84646
S	2.13989	0.4765	0.52876
C	0.96503	-1.45998	-0.79622
H	2.76508	-2.61485	-1.37467
C	0.63976	-0.29957	-0.11534
H	0.20657	-2.08862	-1.27017
C	10.31932	-0.60115	0.24481
C	11.11101	0.29825	0.93282
S	11.35028	-1.85522	-0.55206
C	12.51546	0.01896	0.85816
H	10.69469	1.12986	1.50414
C	12.81865	-1.096	0.11964
H	13.27496	0.62645	1.3564
H	13.79384	-1.5372	-0.08118
C	-0.63976	0.29957	0.11534
S	-2.13989	-0.4765	-0.52876
C	-0.96503	1.45998	0.79622
C	-3.16014	0.82599	0.20162
C	-2.35783	1.75128	0.84646
H	-0.20657	2.08862	1.27017
C	-4.59271	0.74418	0.07242
H	-2.76508	2.61485	1.37467
C	-5.37479	-0.41924	-0.05362
N	-5.44724	1.87318	0.11554
C	-6.72158	0.01413	-0.07886
H	-4.98451	-1.43462	-0.06936
C	-6.7552	1.41361	0.03565
C	-5.07605	3.2499	0.10302

N	-8.0304	-0.44455	-0.15809
C	-8.10267	1.84712	0.01028
C	-4.28423	3.76885	-0.93059
C	-5.53255	4.10807	1.12122
C	-8.88335	0.68491	-0.11504
C	-8.40283	-1.8205	-0.15133
H	-8.49315	2.86233	0.02762
C	-3.92999	5.12512	-0.94468
H	-3.93857	3.10619	-1.72797
C	-5.19865	5.46277	1.10388
H	-6.14732	3.7022	1.92942
C	-10.31932	0.60115	-0.24481
C	-7.92418	-2.68038	-1.15801
C	-9.2174	-2.33819	0.86506
C	-4.39036	5.9768	0.07367
H	-3.30634	5.50207	-1.75777
H	-5.54462	6.14472	1.88388
C	-11.11101	-0.29825	-0.93282
S	-11.35028	1.85522	0.55206
C	-8.25787	-4.03515	-1.14583
H	-7.29187	-2.27585	-1.95325
C	-9.57122	-3.6947	0.87355
H	-9.58001	-1.67469	1.65407
O	-4.10342	7.325	0.14571
C	-12.51546	-0.01896	-0.85816
H	-10.69469	-1.12986	-1.50414
C	-12.81865	1.096	-0.11964
C	-9.08813	-4.54794	-0.13276
H	-7.89451	-4.71809	-1.91703
H	-10.21195	-4.07073	1.67372
C	-3.28448	7.93619	-0.86628
H	-13.27496	-0.62645	-1.3564
H	-13.79384	1.5372	0.08118
O	-9.37245	-5.8969	-0.20852
H	-3.75183	7.8582	-1.86699
H	-3.20404	8.99605	-0.58312
H	-2.27327	7.48606	-0.89515
C	-10.20951	-6.50722	0.78869
H	-10.28246	-7.56798	0.50676
H	-9.76202	-6.42585	1.79821
H	-11.22211	-6.05921	0.79703

Energy Levels

Alpha occ. eigenvalues -- -88.84873 -88.84873 -88.84694 -88.84693 -19.16161

Alpha occ. eigenvalues -- -19.16161 -19.16069 -19.16069 -14.37461 -14.37461
Alpha occ. eigenvalues -- -14.37418 -14.37418 -10.24844 -10.24844 -10.24782
Alpha occ. eigenvalues -- -10.24782 -10.23368 -10.23368 -10.23239 -10.23239
Alpha occ. eigenvalues -- -10.22436 -10.22436 -10.22370 -10.22370 -10.21927
Alpha occ. eigenvalues -- -10.21927 -10.21905 -10.21905 -10.21700 -10.21675
Alpha occ. eigenvalues -- -10.21675 -10.21675 -10.21652 -10.21652 -10.20052
Alpha occ. eigenvalues -- -10.20052 -10.19863 -10.19863 -10.19814 -10.19814
Alpha occ. eigenvalues -- -10.18908 -10.18908 -10.18871 -10.18871 -10.18844
Alpha occ. eigenvalues -- -10.18844 -10.18817 -10.18817 -10.18701 -10.18701
Alpha occ. eigenvalues -- -10.18617 -10.18617 -10.18257 -10.18257 -10.18187
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Alpha occ. eigenvalues -- -10.17679 -10.17679 -10.17666 -10.17666 -10.17097
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Alpha occ. eigenvalues -- -1.06906 -1.06818 -1.06818 -1.00791 -1.00791
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Alpha occ. eigenvalues -- -0.87688 -0.87560 -0.87545 -0.86789 -0.86714
Alpha occ. eigenvalues -- -0.81293 -0.81291 -0.80914 -0.80626 -0.78476
Alpha occ. eigenvalues -- -0.76833 -0.76753 -0.76632 -0.76357 -0.76231
Alpha occ. eigenvalues -- -0.75768 -0.75562 -0.74794 -0.74677 -0.74523
Alpha occ. eigenvalues -- -0.74108 -0.73724 -0.73719 -0.73110 -0.72662
Alpha occ. eigenvalues -- -0.70651 -0.70527 -0.69695 -0.69482 -0.69274
Alpha occ. eigenvalues -- -0.67762 -0.64489 -0.63577 -0.63530 -0.63417
Alpha occ. eigenvalues -- -0.63026 -0.62807 -0.62267 -0.62089 -0.61787
Alpha occ. eigenvalues -- -0.61727 -0.58968 -0.58351 -0.57975 -0.57771
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Alpha occ. eigenvalues -- -0.47811 -0.47641 -0.46520 -0.46319 -0.46230
Alpha occ. eigenvalues -- -0.46128 -0.46127 -0.45648 -0.45583 -0.45247
Alpha occ. eigenvalues -- -0.45228 -0.44729 -0.44725 -0.44299 -0.44296
Alpha occ. eigenvalues -- -0.44229 -0.44209 -0.43550 -0.43502 -0.42991
Alpha occ. eigenvalues -- -0.42871 -0.42778 -0.42702 -0.41626 -0.41439
Alpha occ. eigenvalues -- -0.41318 -0.41251 -0.41072 -0.40995 -0.40673
Alpha occ. eigenvalues -- -0.40560 -0.40294 -0.40136 -0.40108 -0.40012
Alpha occ. eigenvalues -- -0.39874 -0.39694 -0.39465 -0.39323 -0.39063
Alpha occ. eigenvalues -- -0.38522 -0.38439 -0.38212 -0.38164 -0.37791
Alpha occ. eigenvalues -- -0.37589 -0.37478 -0.37308 -0.37291 -0.36754
Alpha occ. eigenvalues -- -0.35942 -0.35463 -0.35029 -0.34460 -0.34457
Alpha occ. eigenvalues -- -0.33501 -0.33448 -0.33353 -0.33352 -0.33278

Alpha occ. eigenvalues -- -0.33265 -0.33147 -0.32976 -0.32622 -0.29937
Alpha occ. eigenvalues -- -0.28844 -0.28423 -0.27515 -0.27441 -0.27392
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Alpha virt. eigenvalues -- 0.05984 0.06071 0.06088 0.06234 0.07065
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Alpha virt. eigenvalues -- 0.09903 0.09905 0.10055 0.10205 0.10325
Alpha virt. eigenvalues -- 0.10428 0.10435 0.10772 0.10900 0.10902
Alpha virt. eigenvalues -- 0.11031 0.11043 0.11114 0.11167 0.11771
Alpha virt. eigenvalues -- 0.11829 0.11992 0.12065 0.12543 0.12566
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Alpha virt. eigenvalues -- 0.12825 0.13210 0.13291 0.13502 0.13705
Alpha virt. eigenvalues -- 0.13819 0.13841 0.14074 0.14214 0.14550
Alpha virt. eigenvalues -- 0.14563 0.14634 0.15017 0.15105 0.15845
Alpha virt. eigenvalues -- 0.16246 0.16314 0.16336 0.16407 0.17209
Alpha virt. eigenvalues -- 0.18976 0.19032 0.19360 0.19593 0.21307
Alpha virt. eigenvalues -- 0.21433 0.21940 0.22259 0.22613 0.22632
Alpha virt. eigenvalues -- 0.22781 0.22783 0.23339 0.23789 0.25082
Alpha virt. eigenvalues -- 0.26051 0.26217 0.26411 0.26733 0.26779
Alpha virt. eigenvalues -- 0.26944 0.27272 0.27893 0.28534 0.28898
Alpha virt. eigenvalues -- 0.28973 0.29063 0.29326 0.29535 0.30107
Alpha virt. eigenvalues -- 0.30291 0.30756 0.30815 0.31131 0.31258
Alpha virt. eigenvalues -- 0.31921 0.31926 0.32144 0.32331 0.32409
Alpha virt. eigenvalues -- 0.32425 0.33339 0.33512 0.33725 0.33790
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Alpha virt. eigenvalues -- 0.36286 0.36449 0.37119 0.37424 0.37565
Alpha virt. eigenvalues -- 0.37697 0.37829 0.38026 0.38238 0.38357
Alpha virt. eigenvalues -- 0.38840 0.39576 0.40186 0.40386 0.40493
Alpha virt. eigenvalues -- 0.40986 0.41143 0.41433 0.41600 0.41734
Alpha virt. eigenvalues -- 0.41779 0.41957 0.42243 0.42776 0.42883
Alpha virt. eigenvalues -- 0.43705 0.44103 0.44594 0.44634 0.44995
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Alpha virt. eigenvalues -- 0.45942 0.45981 0.45983 0.46192 0.46310
Alpha virt. eigenvalues -- 0.46318 0.46340 0.46521 0.46691 0.46913
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Alpha virt. eigenvalues -- 0.47913 0.48106 0.48137 0.48209 0.48438
Alpha virt. eigenvalues -- 0.48802 0.49023 0.49239 0.49406 0.49606

Alpha virt. eigenvalues --	0.49715	0.49943	0.50005	0.50379	0.50729
Alpha virt. eigenvalues --	0.50771	0.50778	0.51602	0.51789	0.52000
Alpha virt. eigenvalues --	0.52178	0.52346	0.52419	0.52874	0.52940
Alpha virt. eigenvalues --	0.52984	0.53285	0.53409	0.53741	0.54085
Alpha virt. eigenvalues --	0.54162	0.54314	0.54521	0.54710	0.54753
Alpha virt. eigenvalues --	0.55508	0.55720	0.55783	0.56012	0.56155
Alpha virt. eigenvalues --	0.56657	0.56833	0.57507	0.57688	0.57779
Alpha virt. eigenvalues --	0.57898	0.58102	0.58172	0.58997	0.59217
Alpha virt. eigenvalues --	0.59426	0.59700	0.59777	0.60256	0.60377
Alpha virt. eigenvalues --	0.60911	0.61188	0.61205	0.61397	0.61480
Alpha virt. eigenvalues --	0.62561	0.62879	0.63070	0.63327	0.63593
Alpha virt. eigenvalues --	0.63762	0.63972	0.64022	0.64333	0.64535
Alpha virt. eigenvalues --	0.64699	0.64976	0.65073	0.65331	0.65365
Alpha virt. eigenvalues --	0.65605	0.65674	0.65682	0.65908	0.66158
Alpha virt. eigenvalues --	0.66251	0.66499	0.66501	0.66754	0.66861
Alpha virt. eigenvalues --	0.66869	0.66886	0.67456	0.67755	0.68442
Alpha virt. eigenvalues --	0.68468	0.68631	0.68632	0.68693	0.68736
Alpha virt. eigenvalues --	0.68974	0.69038	0.69047	0.69340	0.69473
Alpha virt. eigenvalues --	0.69506	0.69860	0.69953	0.70202	0.70377
Alpha virt. eigenvalues --	0.70521	0.70590	0.70597	0.70903	0.71032
Alpha virt. eigenvalues --	0.71317	0.71715	0.71732	0.72299	0.72440
Alpha virt. eigenvalues --	0.72507	0.72806	0.73030	0.73276	0.73497
Alpha virt. eigenvalues --	0.73737	0.73749	0.74209	0.74455	0.74465
Alpha virt. eigenvalues --	0.74606	0.74779	0.74906	0.75080	0.75245
Alpha virt. eigenvalues --	0.75670	0.75734	0.75997	0.76003	0.76156
Alpha virt. eigenvalues --	0.76474	0.76510	0.77049	0.77077	0.77241
Alpha virt. eigenvalues --	0.77309	0.77467	0.77527	0.77598	0.77902
Alpha virt. eigenvalues --	0.78082	0.78653	0.79082	0.79608	0.79776
Alpha virt. eigenvalues --	0.80181	0.80531	0.80610	0.80901	0.81035
Alpha virt. eigenvalues --	0.82119	0.82146	0.82528	0.82551	0.82741
Alpha virt. eigenvalues --	0.82895	0.83244	0.83514	0.83928	0.84018
Alpha virt. eigenvalues --	0.84909	0.85208	0.86123	0.86217	0.86620
Alpha virt. eigenvalues --	0.86807	0.87487	0.87511	0.87560	0.87621
Alpha virt. eigenvalues --	0.89392	0.89395	0.90190	0.90757	0.91464
Alpha virt. eigenvalues --	0.92187	0.92694	0.92834	0.93023	0.93549
Alpha virt. eigenvalues --	0.94168	0.94968	0.95039	0.96103	0.96260
Alpha virt. eigenvalues --	0.96768	0.97143	0.98079	0.98504	0.98555
Alpha virt. eigenvalues --	0.99030	0.99290	0.99488	1.00294	1.01394
Alpha virt. eigenvalues --	1.01418	1.01457	1.01520	1.02143	1.03267
Alpha virt. eigenvalues --	1.03990	1.06834	1.08232	1.09196	1.09745
Alpha virt. eigenvalues --	1.09755	1.09862	1.10626	1.10673	1.11191
Alpha virt. eigenvalues --	1.11210	1.12282	1.12417	1.12757	1.12936
Alpha virt. eigenvalues --	1.13107	1.14253	1.14382	1.14819	1.15165
Alpha virt. eigenvalues --	1.15951	1.16458	1.16622	1.16920	1.17225

Alpha virt. eigenvalues --	1.17312	1.17344	1.17585	1.18501	1.18683
Alpha virt. eigenvalues --	1.19357	1.19378	1.21698	1.22939	1.23205
Alpha virt. eigenvalues --	1.23498	1.23958	1.25056	1.25066	1.25419
Alpha virt. eigenvalues --	1.25508	1.26298	1.27067	1.27553	1.28881
Alpha virt. eigenvalues --	1.31731	1.32044	1.32262	1.34258	1.35728
Alpha virt. eigenvalues --	1.36642	1.36907	1.39282	1.39725	1.41531
Alpha virt. eigenvalues --	1.45986	1.46886	1.48806	1.50766	1.50963
Alpha virt. eigenvalues --	1.51229	1.52218	1.52513	1.54291	1.57785
Alpha virt. eigenvalues --	1.58356	1.58504	1.58518	1.65001	1.67752
Alpha virt. eigenvalues --	1.73569	1.73595	1.73660	1.73662	1.76615
Alpha virt. eigenvalues --	1.77893				

Lowest Excited States (ES)

Table S4. A table of the lowest lying 15 excited states where the energy (eV), oscillator strengths (f), most significant transition, % contribution of the transition are provided for each.

ES	Energy (eV)	*f*	Transition	% Contribution
1	2.37	2.2676	HOMO → LUMO	100%
2	2.84	0.0000	HOMO – 1 → LUMO	65.4%
3	3.17	0.0000	HOMO → LUMO + 1	65.3%
4	3.31	0.0738	HOMO → LUMO + 2	49.6%
5	3.36	0.0855	HOMO – 2 → LUMO	52.5%
6	3.37	0.0000	HOMO – 3 → LUMO	100%
7	3.52	0.1606	HOMO – 1 → LUMO + 1	35.1%
8	3.54	0.0000	HOMO → LUMO + 5	48.7%
9	3.54	0.0000	HOMO → LUMO + 3	46.6%
10	3.55	0.1503	HOMO → LUMO + 6	44.8%
11	3.55	0.1959	HOMO → LUMO + 4	43.9%
12	3.72	0.0000	HOMO → LUMO + 7	62.8%
13	3.76	0.0192	HOMO → LUMO + 8	51.1%
14	3.82	0.0401	HOMO – 4 → LUMO	31.6%
15	3.83	0.0000	HOMO – 1 → LUMO + 2	47.9%

Geometric Images

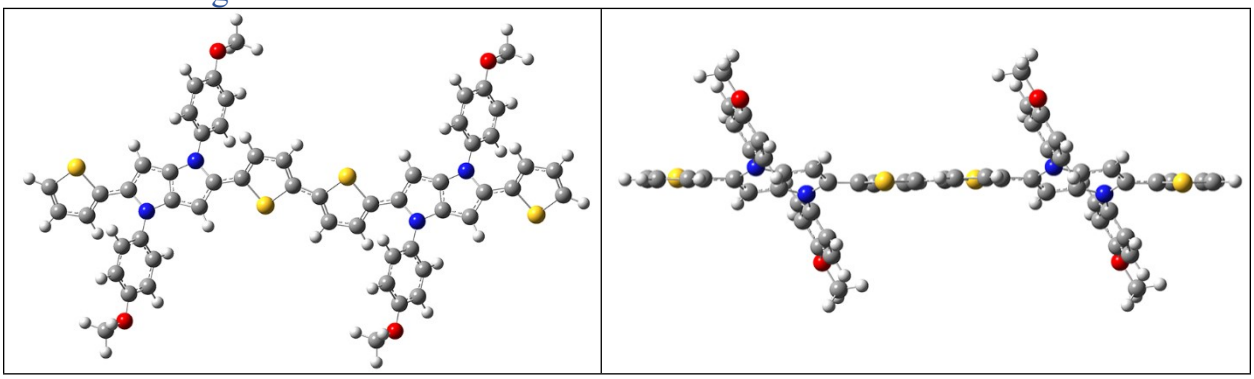

Figure S26. Two geometric perspectives are given with a top view on the left and a front view on the right.

Simulated UV-Vis Spectra

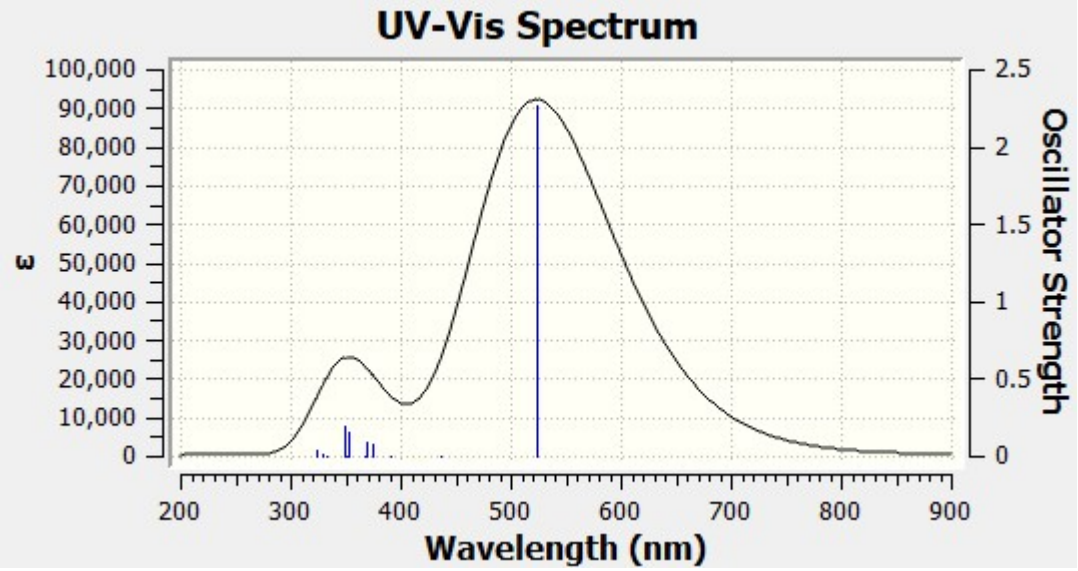

Figure S27. Simulated UV-visible absorbance spectrum of the neutral Th_2DHPP oligomer in gas phase.

Th_2DHPP Radical Cation

Cartesian Coordinates

C	8.834	-0.61746	0.11491
C	8.07523	-1.8128	0.03793
C	6.72962	-1.41716	0.0441
C	6.66298	-0.00005	0.11269
C	4.55398	-0.80591	0.03469
C	5.32585	0.39495	0.09914
H	8.48991	-2.81778	0.01954
H	4.91511	1.40147	0.14522
N	5.44152	-1.91243	-0.0003
N	7.96036	0.48983	0.15292
C	5.12366	-3.30544	-0.11365
C	4.85618	-3.86918	-1.37567
C	5.11029	-4.11789	1.02753
C	4.56161	-5.22677	-1.48699
H	4.87831	-3.23442	-2.26562
C	4.82125	-5.48615	0.92279
H	5.32398	-3.67762	2.00551
C	4.54314	-6.0435	-0.33831
H	4.35041	-5.68728	-2.45451
H	4.81832	-6.1013	1.82467
C	8.29645	1.88211	0.13395
C	8.80775	2.47075	-1.02995

C	8.09147	2.66961	1.28222
C	9.13179	3.83409	-1.05256
H	8.96038	1.85861	-1.92293
C	8.40069	4.02923	1.26356
H	7.69555	2.20684	2.19059
C	8.92653	4.61837	0.09688
H	9.53436	4.27055	-1.96865
H	8.25104	4.6605	2.14215
O	4.24626	-7.36653	-0.5494
O	9.20629	5.96037	0.17668
C	4.22871	-8.29402	0.55494
H	3.97991	-9.27042	0.11526
H	5.21893	-8.3524	1.0451
H	3.45771	-8.01779	1.29915
C	9.7556	6.65701	-0.96
H	9.06246	6.62825	-1.82211
H	9.88828	7.69781	-0.63163
H	10.73585	6.23585	-1.25368
C	3.14	-0.8979	0.02614
C	2.2823	-2.00932	0.06928
S	2.16288	0.62775	-0.03347
C	0.91594	-1.69306	0.06126
H	2.65009	-3.03362	0.11526
C	0.62579	-0.32315	0.0102
H	0.12868	-2.45042	0.09565
C	10.26594	-0.51253	0.174
C	11.08532	0.5185	0.60812
S	11.26511	-1.93027	-0.33865
C	12.48051	0.21409	0.5517
H	10.69842	1.467	0.98225
C	12.74965	-1.04598	0.0754
H	13.26129	0.91129	0.86291
H	13.71719	-1.52511	-0.06946
C	-0.62579	0.32315	-0.0102
S	-2.16288	-0.62775	0.03347
C	-0.91594	1.69306	-0.06125
C	-3.14	0.8979	-0.02613
C	-2.2823	2.00932	-0.06927
H	-0.12868	2.45042	-0.09563
C	-4.55398	0.80591	-0.03468
H	-2.65009	3.03362	-0.11524
C	-5.32585	-0.39495	-0.09914
N	-5.44152	1.91243	0.00031
C	-6.66298	0.00005	-0.11269

H	-4.91511	-1.40147	-0.14523
C	-6.72962	1.41716	-0.0441
C	-5.12366	3.30544	0.11367
N	-7.96036	-0.48983	-0.15293
C	-8.07523	1.8128	-0.03793
C	-5.11029	4.1179	-1.0275
C	-4.85617	3.86917	1.37568
C	-8.834	0.61746	-0.11491
C	-8.29645	-1.88211	-0.13397
H	-8.48991	2.81778	-0.01953
C	-4.82125	5.48615	-0.92276
H	-5.32398	3.67763	-2.00549
C	-4.56161	5.22676	1.48702
H	-4.8783	3.23441	2.26564
C	-10.26594	0.51253	-0.174
C	-8.09146	-2.6696	-1.28224
C	-8.80776	-2.47075	1.02992
C	-4.54314	6.04349	0.33834
H	-4.81832	6.1013	-1.82464
H	-4.3504	5.68727	2.45454
C	-11.08532	-0.51849	-0.60814
S	-11.26511	1.93026	0.33866
C	-8.40068	-4.02922	-1.26358
H	-7.69554	-2.20683	-2.19061
C	-9.1318	-3.83409	1.05252
H	-8.9604	-1.85862	1.92291
O	-4.24626	7.36653	0.54943
C	-12.48051	-0.21409	-0.55172
H	-10.69842	-1.46699	-0.98228
C	-12.74965	1.04598	-0.0754
C	-8.92653	-4.61837	-0.09691
H	-8.25102	-4.66049	-2.14218
H	-9.53437	-4.27056	1.96861
C	-4.22871	8.29402	-0.5549
H	-13.26129	-0.91128	-0.86295
H	-13.71719	1.5251	0.06946
O	-9.20629	-5.96036	-0.17672
H	-5.21893	8.3524	-1.04505
H	-3.97991	9.27042	-0.11522
H	-3.45772	8.0178	-1.29911
C	-9.75561	-6.65701	0.95995
H	-9.88829	-7.69781	0.63158
H	-9.06248	-6.62825	1.82207
H	-10.73586	-6.23585	1.25362

Energy Levels

Alpha occ. eigenvalues -- -88.93533 -88.93532 -88.92292 -88.92292 -19.22616
Alpha occ. eigenvalues -- -19.22616 -19.22213 -19.22213 -14.45964 -14.45964
Alpha occ. eigenvalues -- -14.45703 -14.45703 -10.31933 -10.31933 -10.31594
Alpha occ. eigenvalues -- -10.31594 -10.31541 -10.31541 -10.31469 -10.31469
Alpha occ. eigenvalues -- -10.31337 -10.31337 -10.31131 -10.31097 -10.29717
Alpha occ. eigenvalues -- -10.29717 -10.29504 -10.29504 -10.29345 -10.29345
Alpha occ. eigenvalues -- -10.29239 -10.29239 -10.29191 -10.29191 -10.29115
Alpha occ. eigenvalues -- -10.29115 -10.28773 -10.28773 -10.27565 -10.27565
Alpha occ. eigenvalues -- -10.27271 -10.27268 -10.26947 -10.26947 -10.26508
Alpha occ. eigenvalues -- -10.26508 -10.26304 -10.26304 -10.26059 -10.26059
Alpha occ. eigenvalues -- -10.25804 -10.25804 -10.25720 -10.25720 -10.25634
Alpha occ. eigenvalues -- -10.25634 -10.25518 -10.25518 -10.25408 -10.25408
Alpha occ. eigenvalues -- -10.25178 -10.25178 -10.25111 -10.25111 -10.25000
Alpha occ. eigenvalues -- -10.25000 -10.24743 -10.24743 -8.06205 -8.06204
Alpha occ. eigenvalues -- -8.05041 -8.05041 -6.02669 -6.02669 -6.02231
Alpha occ. eigenvalues -- -6.02231 -6.01595 -6.01594 -6.01462 -6.01462
Alpha occ. eigenvalues -- -6.01018 -6.01018 -6.00517 -6.00517 -1.13497
Alpha occ. eigenvalues -- -1.13497 -1.13077 -1.13077 -1.09603 -1.09603
Alpha occ. eigenvalues -- -1.06488 -1.06487 -0.98875 -0.96842 -0.96155
Alpha occ. eigenvalues -- -0.96155 -0.94893 -0.94886 -0.93948 -0.93906
Alpha occ. eigenvalues -- -0.89705 -0.89282 -0.89072 -0.88901 -0.87501
Alpha occ. eigenvalues -- -0.85525 -0.84059 -0.83750 -0.83532 -0.83387
Alpha occ. eigenvalues -- -0.83226 -0.83176 -0.82835 -0.82787 -0.82002
Alpha occ. eigenvalues -- -0.81632 -0.81602 -0.81374 -0.81225 -0.80933
Alpha occ. eigenvalues -- -0.78144 -0.77309 -0.77308 -0.76465 -0.76379
Alpha occ. eigenvalues -- -0.76094 -0.73186 -0.71655 -0.70633 -0.70542
Alpha occ. eigenvalues -- -0.70289 -0.70261 -0.69479 -0.69459 -0.69257
Alpha occ. eigenvalues -- -0.69169 -0.67910 -0.67147 -0.66525 -0.65274
Alpha occ. eigenvalues -- -0.65082 -0.65018 -0.64063 -0.63550 -0.63543
Alpha occ. eigenvalues -- -0.63007 -0.62821 -0.62359 -0.61602 -0.61044
Alpha occ. eigenvalues -- -0.61042 -0.60273 -0.60229 -0.59843 -0.59684
Alpha occ. eigenvalues -- -0.58132 -0.58127 -0.58095 -0.57921 -0.57720
Alpha occ. eigenvalues -- -0.57118 -0.57103 -0.55958 -0.55305 -0.54180
Alpha occ. eigenvalues -- -0.54122 -0.53992 -0.53950 -0.53671 -0.53671
Alpha occ. eigenvalues -- -0.53494 -0.53475 -0.53123 -0.53096 -0.52979
Alpha occ. eigenvalues -- -0.52946 -0.52565 -0.52564 -0.51847 -0.51438
Alpha occ. eigenvalues -- -0.51217 -0.51205 -0.50826 -0.50824 -0.50372
Alpha occ. eigenvalues -- -0.50112 -0.49729 -0.49670 -0.49656 -0.49361
Alpha occ. eigenvalues -- -0.49123 -0.49100 -0.48779 -0.48669 -0.48611
Alpha occ. eigenvalues -- -0.48172 -0.48170 -0.48060 -0.47779 -0.47544
Alpha occ. eigenvalues -- -0.47534 -0.47465 -0.46741 -0.46733 -0.46437
Alpha occ. eigenvalues -- -0.46219 -0.45894 -0.45726 -0.45635 -0.45316

Alpha occ. eigenvalues -- -0.45230 -0.45106 -0.45001 -0.44288 -0.44167
Alpha occ. eigenvalues -- -0.43661 -0.43427 -0.42791 -0.42100 -0.41730
Alpha occ. eigenvalues -- -0.41729 -0.40314 -0.40146 -0.39924 -0.39785
Alpha occ. eigenvalues -- -0.39693 -0.39653 -0.39349 -0.39296 -0.39192
Alpha occ. eigenvalues -- -0.37776 -0.37264 -0.35588 -0.35068 -0.34816
Alpha occ. eigenvalues -- -0.34326 -0.34301 -0.34169 -0.34069 -0.33209
Alpha occ. eigenvalues -- -0.33167 -0.32726 -0.31460 -0.31404 -0.30767
Alpha occ. eigenvalues -- -0.30691 -0.30302 -0.29602 -0.29568 -0.27030
Alpha occ. eigenvalues -- -0.25067
Alpha virt. eigenvalues -- -0.16815 -0.13324 -0.11375 -0.09591 -0.09342
Alpha virt. eigenvalues -- -0.09187 -0.08918 -0.08907 -0.08618 -0.08516
Alpha virt. eigenvalues -- -0.08034 -0.08019 -0.06813 -0.06347 -0.06332
Alpha virt. eigenvalues -- -0.04288 -0.04083 -0.03239 -0.03020 -0.02824
Alpha virt. eigenvalues -- -0.00987 -0.00205 -0.00196 0.00146 0.00180
Alpha virt. eigenvalues -- 0.00246 0.00250 0.00727 0.01139 0.01379
Alpha virt. eigenvalues -- 0.01572 0.01660 0.01850 0.01874 0.02080
Alpha virt. eigenvalues -- 0.02143 0.02286 0.02308 0.02796 0.03051
Alpha virt. eigenvalues -- 0.03053 0.03892 0.03896 0.04001 0.04061
Alpha virt. eigenvalues -- 0.04248 0.04305 0.04493 0.04698 0.04766
Alpha virt. eigenvalues -- 0.04777 0.05002 0.05350 0.05439 0.05925
Alpha virt. eigenvalues -- 0.06214 0.06219 0.06415 0.06564 0.06583
Alpha virt. eigenvalues -- 0.06697 0.06768 0.06911 0.06979 0.07069
Alpha virt. eigenvalues -- 0.07171 0.07206 0.07499 0.07506 0.07540
Alpha virt. eigenvalues -- 0.08000 0.08011 0.08146 0.08745 0.08878
Alpha virt. eigenvalues -- 0.09293 0.09743 0.09750 0.09976 0.09992
Alpha virt. eigenvalues -- 0.11199 0.11227 0.11321 0.11354 0.13354
Alpha virt. eigenvalues -- 0.13569 0.14136 0.14265 0.14842 0.15025
Alpha virt. eigenvalues -- 0.15488 0.15940 0.16191 0.16304 0.16678
Alpha virt. eigenvalues -- 0.18044 0.18090 0.18782 0.18790 0.19498
Alpha virt. eigenvalues -- 0.19575 0.19878 0.20500 0.21117 0.21404
Alpha virt. eigenvalues -- 0.21962 0.22023 0.22405 0.22647 0.22783
Alpha virt. eigenvalues -- 0.23173 0.23599 0.23676 0.23764 0.23910
Alpha virt. eigenvalues -- 0.24105 0.24340 0.24707 0.24799 0.25327
Alpha virt. eigenvalues -- 0.25458 0.25950 0.26050 0.26200 0.26208
Alpha virt. eigenvalues -- 0.26552 0.26860 0.26914 0.28071 0.28122
Alpha virt. eigenvalues -- 0.28401 0.28710 0.29261 0.29674 0.29885
Alpha virt. eigenvalues -- 0.29938 0.30602 0.30837 0.31503 0.31674
Alpha virt. eigenvalues -- 0.31784 0.32205 0.32527 0.32961 0.33300
Alpha virt. eigenvalues -- 0.33542 0.33559 0.33813 0.33915 0.34204
Alpha virt. eigenvalues -- 0.34848 0.34932 0.35007 0.35184 0.35346
Alpha virt. eigenvalues -- 0.35813 0.36136 0.36719 0.36839 0.37152
Alpha virt. eigenvalues -- 0.37225 0.37619 0.37724 0.38391 0.38580
Alpha virt. eigenvalues -- 0.38763 0.38927 0.38942 0.39102 0.39215
Alpha virt. eigenvalues -- 0.39384 0.39679 0.39762 0.39802 0.39997

Alpha virt. eigenvalues --	0.40138	0.40190	0.40350	0.40612	0.40737
Alpha virt. eigenvalues --	0.40890	0.41103	0.41216	0.41396	0.41403
Alpha virt. eigenvalues --	0.41585	0.41685	0.41731	0.41819	0.42304
Alpha virt. eigenvalues --	0.42550	0.42661	0.42851	0.43218	0.43251
Alpha virt. eigenvalues --	0.43414	0.43416	0.43817	0.43879	0.43966
Alpha virt. eigenvalues --	0.44160	0.44309	0.45293	0.45356	0.45484
Alpha virt. eigenvalues --	0.45917	0.46014	0.46132	0.46158	0.46637
Alpha virt. eigenvalues --	0.46783	0.46949	0.47034	0.47315	0.47691
Alpha virt. eigenvalues --	0.47737	0.48348	0.48571	0.48611	0.48808
Alpha virt. eigenvalues --	0.49236	0.49329	0.49438	0.49855	0.50394
Alpha virt. eigenvalues --	0.50472	0.50951	0.51015	0.51365	0.51421
Alpha virt. eigenvalues --	0.51740	0.51812	0.52434	0.52524	0.53003
Alpha virt. eigenvalues --	0.53094	0.53233	0.53712	0.53983	0.54502
Alpha virt. eigenvalues --	0.54663	0.54847	0.55493	0.56059	0.56398
Alpha virt. eigenvalues --	0.57080	0.57302	0.57910	0.58092	0.58207
Alpha virt. eigenvalues --	0.58256	0.58770	0.58782	0.58993	0.59013
Alpha virt. eigenvalues --	0.59151	0.59183	0.59361	0.59506	0.59680
Alpha virt. eigenvalues --	0.59792	0.60322	0.60589	0.60699	0.60733
Alpha virt. eigenvalues --	0.60801	0.61098	0.61106	0.61260	0.61409
Alpha virt. eigenvalues --	0.61419	0.61553	0.61739	0.61847	0.61859
Alpha virt. eigenvalues --	0.62052	0.62233	0.62438	0.62478	0.62592
Alpha virt. eigenvalues --	0.62664	0.62752	0.63034	0.63042	0.63171
Alpha virt. eigenvalues --	0.63295	0.63335	0.63727	0.63756	0.63915
Alpha virt. eigenvalues --	0.64237	0.64365	0.64788	0.64800	0.64811
Alpha virt. eigenvalues --	0.65125	0.65151	0.65265	0.65547	0.65721
Alpha virt. eigenvalues --	0.65734	0.65990	0.66086	0.66406	0.66716
Alpha virt. eigenvalues --	0.66745	0.67005	0.67153	0.67386	0.67422
Alpha virt. eigenvalues --	0.67582	0.67807	0.68130	0.68222	0.68317
Alpha virt. eigenvalues --	0.68454	0.68574	0.68759	0.69089	0.69184
Alpha virt. eigenvalues --	0.69211	0.69223	0.69656	0.69753	0.69967
Alpha virt. eigenvalues --	0.70056	0.70318	0.70688	0.70892	0.71244
Alpha virt. eigenvalues --	0.71687	0.72239	0.72363	0.73019	0.73219
Alpha virt. eigenvalues --	0.73428	0.73498	0.74598	0.74932	0.75256
Alpha virt. eigenvalues --	0.75390	0.75401	0.75786	0.76512	0.76814
Alpha virt. eigenvalues --	0.76830	0.77121	0.77466	0.77593	0.78714
Alpha virt. eigenvalues --	0.78788	0.79059	0.80235	0.80282	0.80352
Alpha virt. eigenvalues --	0.81745	0.81956	0.82907	0.82959	0.84288
Alpha virt. eigenvalues --	0.84377	0.85770	0.85862	0.87172	0.87377
Alpha virt. eigenvalues --	0.88665	0.88956	0.89217	0.89669	0.89919
Alpha virt. eigenvalues --	0.90227	0.90528	0.91005	0.91847	0.91848
Alpha virt. eigenvalues --	0.92487	0.93036	0.93436	0.93862	0.93868
Alpha virt. eigenvalues --	0.94623	0.95925	0.96824	0.97588	0.97593
Alpha virt. eigenvalues --	0.98605	0.99295	1.01119	1.01317	1.02216
Alpha virt. eigenvalues --	1.03415	1.03763	1.03923	1.04243	1.04464

Alpha virt. eigenvalues --	1.04522	1.04735	1.04823	1.05502	1.06112
Alpha virt. eigenvalues --	1.06209	1.06615	1.06622	1.08031	1.08231
Alpha virt. eigenvalues --	1.08310	1.08343	1.09619	1.09822	1.10897
Alpha virt. eigenvalues --	1.10998	1.11468	1.11613	1.12519	1.12629
Alpha virt. eigenvalues --	1.12792	1.12978	1.15113	1.15445	1.16779
Alpha virt. eigenvalues --	1.17267	1.18155	1.18413	1.19018	1.19063
Alpha virt. eigenvalues --	1.19546	1.19883	1.20784	1.20800	1.22100
Alpha virt. eigenvalues --	1.22210	1.24403	1.24450	1.27731	1.29447
Alpha virt. eigenvalues --	1.29492	1.30099	1.33913	1.34064	1.35434
Alpha virt. eigenvalues --	1.38138	1.39295	1.41703	1.43312	1.44514
Alpha virt. eigenvalues --	1.45270	1.47474	1.47802	1.48157	1.52026
Alpha virt. eigenvalues --	1.52477	1.53160	1.53234	1.57485	1.60286
Alpha virt. eigenvalues --	1.67344	1.67354	1.67498	1.67513	1.69422
Alpha virt. eigenvalues --	1.70978				

Lowest Excited States

Table S5. A table of the lowest lying 15 excited states where the energy (eV), oscillator strengths (f), most significant transition, % contribution of the transition are provided for each.

ES	Energy (eV)	*f*	Transition	% Contribution
1	0.85	0.6334	$SOMO_\beta \rightarrow LUMO_\beta$	62.8%
2	1.60	0.0000	$SOMO_\beta - 1 \rightarrow LUMO_\beta$	58.1%
3	1.61	0.0038	$SOMO_\beta - 2 \rightarrow LUMO_\beta$	71.5%
4	1.66	0.0000	$SOMO_\beta - 3 \rightarrow LUMO_\beta$	45.3%
5	1.72	2.0932	$SOMO_\alpha \rightarrow LUMO_\alpha$	50.6%
6	1.85	0.0000	$SOMO_\beta \rightarrow LUMO_\beta + 1$	42.5%
7	2.02	0.0003	$SOMO_\beta - 4 \rightarrow LUMO_\beta$	78.6%
8	2.03	0.0000	$SOMO_\beta - 5 \rightarrow LUMO_\beta$	89.0%
9	2.11	0.0027	$SOMO_\beta - 6 \rightarrow LUMO_\beta$	68.8%
10	2.11	0.0000	$SOMO_\beta - 7 \rightarrow LUMO_\beta$	64.4%
11	2.27	0.0061	$SOMO_\beta - 8 \rightarrow LUMO_\beta$	34.8%
12	2.51	0.0000	$SOMO_\alpha - 1 \rightarrow LUMO_\alpha$	34.1%
13	2.54	0.0051	$SOMO_\beta - 8 \rightarrow LUMO_\beta$	23.0%
14	2.62	0.0000	$SOMO_\beta - 8 \rightarrow LUMO_\beta$	70.7%
15	2.65	0.0035	$SOMO_\beta - 10 \rightarrow LUMO_\beta$	53.7%

Geometric Images

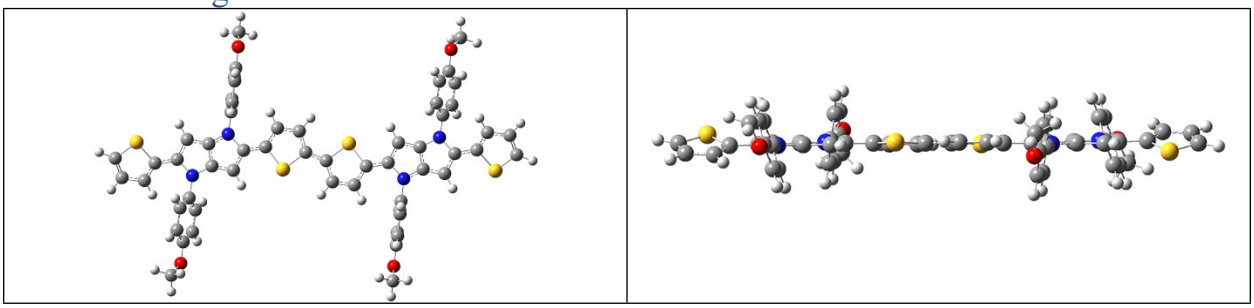

Figure S28. Two geometric perspectives are given with a top view on the left and a front view on the right.

Simulated UV-Vis Spectra

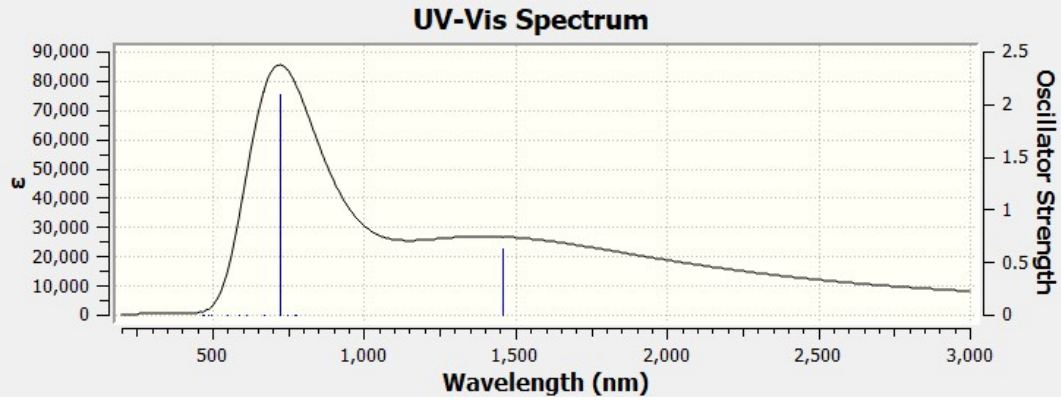

Figure S29. Simulated UV-visible absorbance spectrum of the neutral Th_2DHPP+1 oligomer in gas phase.

BTD_2DHPP Neutral

Cartesian Coordinates

C	0.96003	3.08174	0.00794
C	1.11811	1.60909	0.00325
C	-0.04628	0.73395	-0.00161
C	-1.26997	1.40247	-0.01405
C	-1.41919	2.81405	-0.01388
C	-0.35083	3.70452	0.00921
H	-2.17604	0.79478	-0.03269
H	-2.43775	3.20258	-0.05617
S	3.4207	2.62119	0.01405
N	2.39014	1.20644	0.00609
N	2.10082	3.76719	0.01373
C	-0.49676	5.14878	-0.00249
C	0.41321	6.09729	-0.51156
N	-1.67975	5.82353	0.40946
C	-0.23028	7.3507	-0.42494
H	1.39618	5.85365	-0.90075
C	-1.5086	7.17284	0.13325
C	-2.76375	5.31875	1.18784
N	-0.05576	8.7044	-0.68683
C	-2.14399	8.43274	0.23679
C	-2.53221	4.69847	2.42328
C	-4.08623	5.47634	0.73186
C	-1.22537	9.37078	-0.26946
C	1.04923	9.25265	-1.39862
H	-3.10879	8.6776	0.67515

C	-3.60165	4.21645	3.19077
H	-1.50682	4.58919	2.78583
C	-5.15663	5.01633	1.49957
H	-4.26665	5.95516	-0.23462
C	2.36203	8.95251	-0.99081
C	0.84628	10.06486	-2.52119
C	-4.91933	4.3776	2.73031
H	-3.39272	3.73001	4.14568
H	-6.18951	5.12908	1.16248
C	3.45072	9.46081	-1.69637
H	2.52038	8.33437	-0.10367
C	1.93811	10.59958	-3.22195
H	-0.17122	10.28019	-2.85799
O	-6.03803	3.94594	3.41301
C	3.24519	10.29425	-2.81127
H	4.47696	9.24072	-1.39375
H	1.75174	11.23489	-4.0903
C	-5.88967	3.27301	4.6756
O	4.38543	10.76121	-3.43656
H	-5.4095	3.92713	5.42889
H	-6.91073	3.02823	5.00329
H	-5.30419	2.33928	4.56972
C	4.26755	11.62327	-4.58046
H	5.29853	11.85782	-4.88417
H	3.74507	11.11875	-5.41646
H	3.73709	12.56194	-4.32756
C	-1.41815	10.81848	-0.31572
C	-0.41084	11.751	0.14546
C	-2.62558	11.36244	-0.73133
C	-0.67576	13.19455	0.12365
N	0.77476	11.39903	0.64054
C	-2.88359	12.773	-0.73578
H	-3.4045	10.68465	-1.09221
C	-1.93811	13.6878	-0.33398
N	0.31803	13.96235	0.58077
S	1.61436	12.88041	1.07029
H	-3.86054	13.12045	-1.08531
H	-2.11635	14.76457	-0.34239
C	0.04628	-0.73395	-0.00161
C	-1.11811	-1.60909	0.00325
C	1.26997	-1.40247	-0.01405
C	-0.96003	-3.08174	0.00794
N	-2.39014	-1.20644	0.00609
C	1.41919	-2.81405	-0.01388

H	2.17604	-0.79478	-0.03269
C	0.35083	-3.70452	0.00921
N	-2.10082	-3.76719	0.01373
S	-3.4207	-2.62119	0.01405
H	2.43775	-3.20258	-0.05617
C	0.49676	-5.14878	-0.00249
C	-0.41321	-6.09729	-0.51156
N	1.67975	-5.82353	0.40946
C	0.23028	-7.3507	-0.42494
H	-1.39618	-5.85365	-0.90075
C	1.5086	-7.17284	0.13325
C	2.76375	-5.31875	1.18784
N	0.05576	-8.7044	-0.68683
C	2.14399	-8.43274	0.23679
C	2.53221	-4.69847	2.42328
C	4.08623	-5.47634	0.73186
C	1.22537	-9.37078	-0.26946
C	-1.04923	-9.25265	-1.39862
H	3.10879	-8.6776	0.67515
C	3.60165	-4.21645	3.19077
H	1.50682	-4.58919	2.78583
C	5.15663	-5.01633	1.49957
H	4.26665	-5.95516	-0.23462
C	1.41815	-10.81848	-0.31572
C	-2.36203	-8.95251	-0.99081
C	-0.84628	-10.06486	-2.52119
C	4.91933	-4.3776	2.73031
H	3.39272	-3.73001	4.14568
H	6.18951	-5.12908	1.16248
C	0.41084	-11.751	0.14546
C	2.62558	-11.36244	-0.73133
C	-3.45072	-9.46081	-1.69637
H	-2.52038	-8.33437	-0.10367
C	-1.93811	-10.59958	-3.22195
H	0.17122	-10.28019	-2.85799
O	6.03803	-3.94594	3.41301
C	0.67576	-13.19455	0.12365
N	-0.77476	-11.39903	0.64054
C	2.88359	-12.773	-0.73578
H	3.4045	-10.68465	-1.09221
C	-3.24519	-10.29425	-2.81127
H	-4.47696	-9.24072	-1.39375
H	-1.75174	-11.23489	-4.0903
C	5.88967	-3.27301	4.6756

C	1.93811	-13.6878	-0.33398
N	-0.31803	-13.96235	0.58077
S	-1.61436	-12.88041	1.07029
H	3.86054	-13.12045	-1.08531
O	-4.38543	-10.76121	-3.43656
H	5.4095	-3.92713	5.42889
H	6.91073	-3.02823	5.00329
H	5.30419	-2.33928	4.56972
H	2.11635	-14.76457	-0.34239
C	-4.26755	-11.62327	-4.58046
H	-5.29853	-11.85782	-4.88417
H	-3.74507	-11.11875	-5.41646
H	-3.73709	-12.56194	-4.32756

Energy Levels

Alpha occ. eigenvalues -- -88.90561 -88.90561 -88.90503 -88.90502 -19.16321
Alpha occ. eigenvalues -- -19.16321 -19.15669 -19.15669 -14.37752 -14.37752
Alpha occ. eigenvalues -- -14.37629 -14.37629 -14.36856 -14.36856 -14.36669
Alpha occ. eigenvalues -- -14.36669 -14.36663 -14.36663 -14.36529 -14.36527
Alpha occ. eigenvalues -- -10.25074 -10.25074 -10.24250 -10.24250 -10.24212
Alpha occ. eigenvalues -- -10.24210 -10.24164 -10.24163 -10.24142 -10.24142
Alpha occ. eigenvalues -- -10.23918 -10.23918 -10.23448 -10.23448 -10.22972
Alpha occ. eigenvalues -- -10.22972 -10.22751 -10.22751 -10.22239 -10.22239
Alpha occ. eigenvalues -- -10.22072 -10.22072 -10.21929 -10.21928 -10.21633
Alpha occ. eigenvalues -- -10.21633 -10.20889 -10.20888 -10.20377 -10.20357
Alpha occ. eigenvalues -- -10.20342 -10.20342 -10.20068 -10.20068 -10.20052
Alpha occ. eigenvalues -- -10.20052 -10.19794 -10.19794 -10.19683 -10.19683
Alpha occ. eigenvalues -- -10.19178 -10.19178 -10.19165 -10.19165 -10.18952
Alpha occ. eigenvalues -- -10.18952 -10.18536 -10.18536 -10.18498 -10.18498
Alpha occ. eigenvalues -- -10.18430 -10.18430 -10.18263 -10.18263 -10.18153
Alpha occ. eigenvalues -- -10.18153 -10.17823 -10.17822 -10.17656 -10.17656
Alpha occ. eigenvalues -- -10.17644 -10.17644 -10.17032 -10.17032 -8.03185
Alpha occ. eigenvalues -- -8.03185 -8.03126 -8.03125 -5.99691 -5.99691
Alpha occ. eigenvalues -- -5.99645 -5.99645 -5.99133 -5.99133 -5.99086
Alpha occ. eigenvalues -- -5.99086 -5.98582 -5.98582 -5.98499 -5.98499
Alpha occ. eigenvalues -- -1.07078 -1.07078 -1.06401 -1.06401 -1.02160
Alpha occ. eigenvalues -- -1.02119 -1.02112 -1.02086 -1.01039 -1.01039
Alpha occ. eigenvalues -- -0.98201 -0.98201 -0.90699 -0.90404 -0.90403
Alpha occ. eigenvalues -- -0.90373 -0.87659 -0.87642 -0.87279 -0.87214
Alpha occ. eigenvalues -- -0.87142 -0.86556 -0.86469 -0.85570 -0.81389
Alpha occ. eigenvalues -- -0.81388 -0.81175 -0.80900 -0.79620 -0.78224
Alpha occ. eigenvalues -- -0.77599 -0.77113 -0.77105 -0.77097 -0.76870
Alpha occ. eigenvalues -- -0.76738 -0.76487 -0.76169 -0.75992 -0.75750
Alpha occ. eigenvalues -- -0.75159 -0.74892 -0.74482 -0.74289 -0.71541

Alpha occ. eigenvalues -- -0.70730 -0.70700 -0.69931 -0.69445 -0.69419
Alpha occ. eigenvalues -- -0.67399 -0.66534 -0.66121 -0.65438 -0.64857
Alpha occ. eigenvalues -- -0.64153 -0.63784 -0.63741 -0.63151 -0.62991
Alpha occ. eigenvalues -- -0.62531 -0.62481 -0.62192 -0.61812 -0.61719
Alpha occ. eigenvalues -- -0.61577 -0.61408 -0.61134 -0.60159 -0.59083
Alpha occ. eigenvalues -- -0.58187 -0.58044 -0.57781 -0.57652 -0.56385
Alpha occ. eigenvalues -- -0.56333 -0.55200 -0.54793 -0.54428 -0.54268
Alpha occ. eigenvalues -- -0.53684 -0.53653 -0.52360 -0.52243 -0.51536
Alpha occ. eigenvalues -- -0.51315 -0.51128 -0.51094 -0.50806 -0.50778
Alpha occ. eigenvalues -- -0.50576 -0.50135 -0.49756 -0.49733 -0.49532
Alpha occ. eigenvalues -- -0.48937 -0.48767 -0.48452 -0.48201 -0.48151
Alpha occ. eigenvalues -- -0.48039 -0.48001 -0.47501 -0.47493 -0.47352
Alpha occ. eigenvalues -- -0.46506 -0.46497 -0.46095 -0.46074 -0.45991
Alpha occ. eigenvalues -- -0.45742 -0.45549 -0.45496 -0.45457 -0.45163
Alpha occ. eigenvalues -- -0.44884 -0.44704 -0.44611 -0.44583 -0.44478
Alpha occ. eigenvalues -- -0.44382 -0.44014 -0.43956 -0.43774 -0.43528
Alpha occ. eigenvalues -- -0.43468 -0.43194 -0.42683 -0.42628 -0.41806
Alpha occ. eigenvalues -- -0.41447 -0.40944 -0.40800 -0.40761 -0.40497
Alpha occ. eigenvalues -- -0.40454 -0.40443 -0.40092 -0.39819 -0.39781
Alpha occ. eigenvalues -- -0.39769 -0.39546 -0.39541 -0.39402 -0.39390
Alpha occ. eigenvalues -- -0.39358 -0.39159 -0.38665 -0.38554 -0.38267
Alpha occ. eigenvalues -- -0.38217 -0.38110 -0.37666 -0.37488 -0.37478
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Alpha occ. eigenvalues -- -0.33512 -0.33492 -0.33386 -0.33343 -0.32930
Alpha occ. eigenvalues -- -0.32925 -0.32545 -0.32408 -0.32340 -0.31796
Alpha occ. eigenvalues -- -0.31768 -0.30931 -0.29480 -0.29479 -0.29428
Alpha occ. eigenvalues -- -0.29022 -0.28586 -0.28365 -0.28295 -0.28294
Alpha occ. eigenvalues -- -0.28124 -0.28004 -0.27549 -0.27494 -0.27218
Alpha occ. eigenvalues -- -0.26771 -0.26715 -0.26005 -0.25243 -0.25130
Alpha occ. eigenvalues -- -0.24952 -0.23412 -0.23086 -0.22996 -0.20973
Alpha occ. eigenvalues -- -0.20856 -0.19674 -0.17921
Alpha virt. eigenvalues -- -0.11460 -0.10629 -0.10383 -0.09385 -0.04464
Alpha virt. eigenvalues -- -0.03499 -0.02672 -0.02543 -0.01972 -0.01820
Alpha virt. eigenvalues -- -0.01742 -0.01723 -0.01651 -0.01606 -0.01550
Alpha virt. eigenvalues -- -0.01516 -0.01335 -0.00244 -0.00238 -0.00084
Alpha virt. eigenvalues -- -0.00012 0.00192 0.01159 0.01173 0.03565
Alpha virt. eigenvalues -- 0.04042 0.04051 0.04417 0.04705 0.05074
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Alpha virt. eigenvalues -- 0.06702 0.07096 0.07479 0.07610 0.07876
Alpha virt. eigenvalues -- 0.08268 0.08606 0.09429 0.09466 0.10143
Alpha virt. eigenvalues -- 0.10197 0.10443 0.10444 0.10618 0.10709
Alpha virt. eigenvalues -- 0.10880 0.10923 0.10955 0.11033 0.11106
Alpha virt. eigenvalues -- 0.11254 0.11910 0.11933 0.12227 0.12234

Alpha virt. eigenvalues -- 0.12512 0.12572 0.12624 0.12631 0.13013
Alpha virt. eigenvalues -- 0.13013 0.13086 0.13146 0.13316 0.13361
Alpha virt. eigenvalues -- 0.13818 0.13887 0.14072 0.14074 0.14218
Alpha virt. eigenvalues -- 0.14405 0.14603 0.14650 0.14802 0.15334
Alpha virt. eigenvalues -- 0.15360 0.15771 0.15798 0.16413 0.16462
Alpha virt. eigenvalues -- 0.16892 0.16953 0.17949 0.18389 0.18483
Alpha virt. eigenvalues -- 0.19294 0.19816 0.20164 0.20270 0.20948
Alpha virt. eigenvalues -- 0.21075 0.21810 0.22023 0.22171 0.22343
Alpha virt. eigenvalues -- 0.22378 0.22451 0.22642 0.22860 0.23151
Alpha virt. eigenvalues -- 0.23193 0.23591 0.24229 0.24405 0.25785
Alpha virt. eigenvalues -- 0.25832 0.26059 0.26167 0.26379 0.26637
Alpha virt. eigenvalues -- 0.26744 0.27153 0.27440 0.27480 0.28444
Alpha virt. eigenvalues -- 0.28558 0.28875 0.28894 0.29102 0.29113
Alpha virt. eigenvalues -- 0.29287 0.29467 0.29708 0.29934 0.30064
Alpha virt. eigenvalues -- 0.30388 0.30633 0.30849 0.30977 0.31038
Alpha virt. eigenvalues -- 0.31152 0.31566 0.31698 0.31764 0.31994
Alpha virt. eigenvalues -- 0.32073 0.32513 0.32762 0.32844 0.32858
Alpha virt. eigenvalues -- 0.33207 0.34143 0.34376 0.34560 0.34733
Alpha virt. eigenvalues -- 0.35065 0.35257 0.35467 0.35491 0.35943
Alpha virt. eigenvalues -- 0.36172 0.36228 0.36780 0.37370 0.37377
Alpha virt. eigenvalues -- 0.37744 0.37809 0.38025 0.38161 0.38545
Alpha virt. eigenvalues -- 0.38837 0.39006 0.39039 0.39453 0.40153
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Alpha virt. eigenvalues -- 0.41271 0.41426 0.41633 0.42199 0.43221
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Alpha virt. eigenvalues -- 0.44386 0.44657 0.44903 0.44917 0.45164
Alpha virt. eigenvalues -- 0.45199 0.45288 0.45333 0.45747 0.45764
Alpha virt. eigenvalues -- 0.45986 0.46111 0.46119 0.46196 0.46379
Alpha virt. eigenvalues -- 0.46427 0.46583 0.46686 0.46700 0.46734
Alpha virt. eigenvalues -- 0.47410 0.47527 0.47840 0.47857 0.47956
Alpha virt. eigenvalues -- 0.47962 0.48380 0.48499 0.48502 0.48528
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Alpha virt. eigenvalues -- 0.49710 0.49786 0.50073 0.50274 0.50444
Alpha virt. eigenvalues -- 0.50768 0.51197 0.51348 0.51518 0.51751
Alpha virt. eigenvalues -- 0.51915 0.52362 0.52708 0.52739 0.52850
Alpha virt. eigenvalues -- 0.52881 0.53016 0.53305 0.53405 0.54004
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Alpha virt. eigenvalues -- 0.55208 0.55630 0.55833 0.56003 0.56099
Alpha virt. eigenvalues -- 0.56288 0.56290 0.56592 0.56698 0.57079
Alpha virt. eigenvalues -- 0.57151 0.57401 0.57664 0.57837 0.58016
Alpha virt. eigenvalues -- 0.58059 0.58710 0.58812 0.59006 0.59186
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Alpha virt. eigenvalues -- 0.60376 0.61108 0.61337 0.61411 0.61588
Alpha virt. eigenvalues -- 0.61753 0.62084 0.62368 0.62435 0.62582

Alpha virt. eigenvalues -- 0.62859 0.63385 0.63508 0.63515 0.63620
Alpha virt. eigenvalues -- 0.63995 0.64215 0.64231 0.64238 0.64581
Alpha virt. eigenvalues -- 0.64593 0.64840 0.65296 0.65312 0.65380
Alpha virt. eigenvalues -- 0.65488 0.65542 0.65600 0.65944 0.65953
Alpha virt. eigenvalues -- 0.66156 0.66200 0.66330 0.66571 0.66647
Alpha virt. eigenvalues -- 0.66663 0.66731 0.66826 0.67157 0.67207
Alpha virt. eigenvalues -- 0.67229 0.67419 0.67576 0.67594 0.67871
Alpha virt. eigenvalues -- 0.67931 0.68212 0.68285 0.68414 0.68648
Alpha virt. eigenvalues -- 0.68681 0.68841 0.68853 0.68888 0.68960
Alpha virt. eigenvalues -- 0.69375 0.69473 0.69548 0.70031 0.70049
Alpha virt. eigenvalues -- 0.70128 0.70167 0.70489 0.70628 0.70668
Alpha virt. eigenvalues -- 0.70830 0.70912 0.70931 0.71067 0.71085
Alpha virt. eigenvalues -- 0.71197 0.71400 0.71962 0.72031 0.72206
Alpha virt. eigenvalues -- 0.72583 0.72813 0.72874 0.72919 0.72981
Alpha virt. eigenvalues -- 0.73195 0.73200 0.73793 0.73995 0.74101
Alpha virt. eigenvalues -- 0.74161 0.74333 0.74744 0.74934 0.75202
Alpha virt. eigenvalues -- 0.75516 0.75724 0.75898 0.75940 0.76284
Alpha virt. eigenvalues -- 0.76323 0.76480 0.76514 0.77018 0.77424
Alpha virt. eigenvalues -- 0.77763 0.77921 0.78103 0.78368 0.78622
Alpha virt. eigenvalues -- 0.78737 0.78880 0.79147 0.79386 0.79467
Alpha virt. eigenvalues -- 0.79767 0.79896 0.80187 0.80319 0.80692
Alpha virt. eigenvalues -- 0.80790 0.80908 0.81061 0.81263 0.81439
Alpha virt. eigenvalues -- 0.81963 0.82250 0.82722 0.83000 0.83373
Alpha virt. eigenvalues -- 0.83525 0.84244 0.84344 0.84348 0.84497
Alpha virt. eigenvalues -- 0.84770 0.85030 0.85125 0.85344 0.85656
Alpha virt. eigenvalues -- 0.85674 0.86084 0.86535 0.86695 0.87094
Alpha virt. eigenvalues -- 0.87252 0.87697 0.87786 0.88535 0.88619
Alpha virt. eigenvalues -- 0.89574 0.90052 0.90101 0.90427 0.90826
Alpha virt. eigenvalues -- 0.91122 0.91323 0.91469 0.91937 0.91978
Alpha virt. eigenvalues -- 0.92403 0.92753 0.93569 0.93714 0.94195
Alpha virt. eigenvalues -- 0.94261 0.94899 0.95059 0.96214 0.96353
Alpha virt. eigenvalues -- 0.97293 0.97612 0.98025 0.98255 0.98856
Alpha virt. eigenvalues -- 0.98856 1.00169 1.00351 1.01366 1.01380
Alpha virt. eigenvalues -- 1.01481 1.01722 1.02195 1.02317 1.03361
Alpha virt. eigenvalues -- 1.03415 1.05575 1.06059 1.07330 1.07461
Alpha virt. eigenvalues -- 1.07782 1.08316 1.09396 1.09685 1.10200
Alpha virt. eigenvalues -- 1.10270 1.10540 1.10734 1.10795 1.11057
Alpha virt. eigenvalues -- 1.11311 1.11337 1.12285 1.12315 1.12754
Alpha virt. eigenvalues -- 1.13105 1.13341 1.13700 1.14470 1.14750
Alpha virt. eigenvalues -- 1.15111 1.15331 1.16204 1.16524 1.16893
Alpha virt. eigenvalues -- 1.16910 1.17475 1.17584 1.17977 1.18197
Alpha virt. eigenvalues -- 1.18489 1.18497 1.19358 1.19720 1.20458
Alpha virt. eigenvalues -- 1.20704 1.21054 1.21591 1.22354 1.22514
Alpha virt. eigenvalues -- 1.23339 1.23652 1.24710 1.24815 1.25006

Alpha virt. eigenvalues --	1.25454	1.27056	1.27659	1.29151	1.29464
Alpha virt. eigenvalues --	1.29848	1.29988	1.30527	1.30576	1.31253
Alpha virt. eigenvalues --	1.31676	1.32224	1.32647	1.33109	1.34155
Alpha virt. eigenvalues --	1.35958	1.36545	1.38617	1.38651	1.41521
Alpha virt. eigenvalues --	1.41542	1.42279	1.42324	1.43259	1.43444
Alpha virt. eigenvalues --	1.44779	1.47077	1.47980	1.48824	1.49091
Alpha virt. eigenvalues --	1.49966	1.50654	1.50691	1.53032	1.53119
Alpha virt. eigenvalues --	1.53153	1.54420	1.56093	1.56133	1.57486
Alpha virt. eigenvalues --	1.58142	1.58763	1.58780	1.60318	1.63673
Alpha virt. eigenvalues --	1.68681	1.68811	1.73495	1.73496	1.74358
Alpha virt. eigenvalues --	1.74362	1.78168	1.78209		

Lowest Excited States

Table S6. A table of the lowest lying 15 excited states where the energy (eV), oscillator strengths (f), most significant transition, % contribution of the transition are provided for each.

ES	Energy (eV)	*f*	Transition	% Contribution
1	1.48	1.3047	HOMO → LUMO	100%
2	1.72	0.0000	HOMO → LUMO + 1	84.2%
3	1.77	0.0100	HOMO → LUMO + 2	100%
4	1.89	0.0000	HOMO → LUMO + 3	83.3%
5	1.97	0.0011	HOMO – 1 → LUMO	73.0%
6	2.14	0.0048	HOMO – 2 → LUMO	54.1%
7	2.18	0.0006	HOMO – 3 → LUMO	45.6%
8	2.20	0.0348	HOMO – 1 → LUMO + 1	57.2%
9	2.23	0.0007	HOMO – 1 → LUMO + 2	47.1%
10	2.32	0.0059	HOMO – 2 → LUMO + 2	31.8%
11	2.34	0.0003	HOMO – 3 → LUMO + 2	30.9%
12	2.43	0.0026	HOMO – 1 → LUMO + 3	100%
13	2.64	0.0002	HOMO – 3 → LUMO + 2 HOMO – 2 → LUMO + 1	33.5% 34.1%
14	2.65	0.0010	HOMO – 3 → LUMO + 1	37.8%
15	2.78	0.0001	HOMO – 2 → LUMO + 3	60.9%

Geometric Images

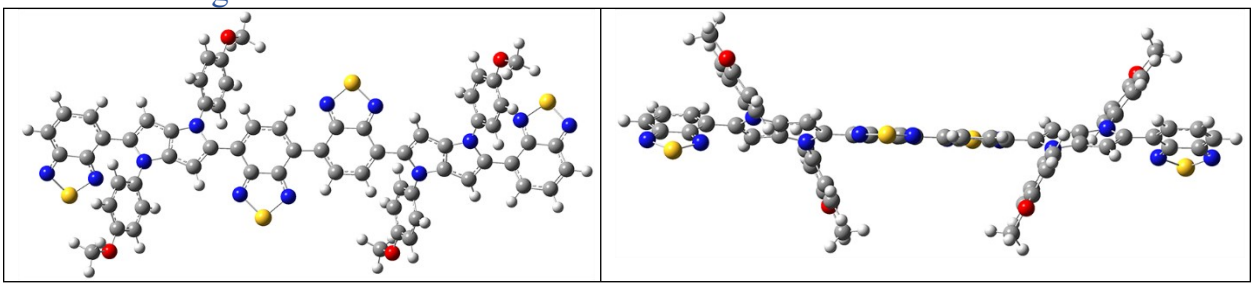

Figure S30. Two geometric perspectives are given with a top view on the left and a front view on the right.

Simulated UV-Vis Spectra

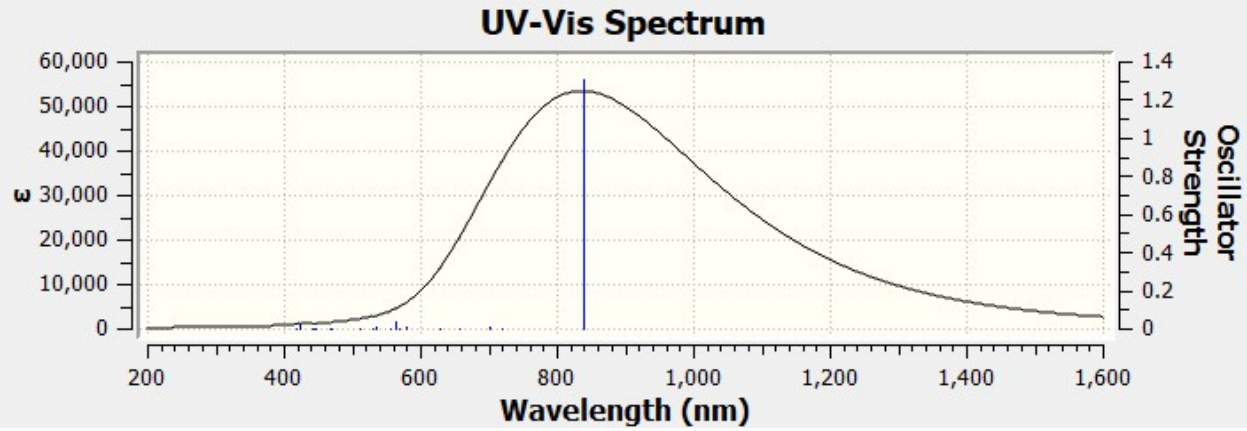

Figure S31. Simulated UV-visible absorbance spectrum of the neutral BDT_2DHPP oligomer in gas phase.

References

(1) Bartlett, K. A.; Charland-Martin, A.; Lawton, J.; Tomlinson, A. L.; Collier, G. S. Azomethine-Containing Pyrrolo[3,2-b]Pyrrole Copolymers for Simple and Degradable Conjugated Polymers. *Macromol Rapid Commun* **2024**, *45* (1), 2300220. https://doi.org/10.1002/marc.202300220.

(2) Bell, K.-J. J.; Sabury, S.; Phan, V.; Wagner, E. M.; Hawks, A. M.; Bartlett, K. A.; Collier, G. S. Synthesis of 1,4-Dihydropyrrolo[3,2-b]Pyrrole-Containing Donor–Acceptor Copolymers and Their Optoelectronic Properties. *J Poly Sci* **2024**, *62*, 2975. https://doi.org/10.1002/pol.20240093.