

Supporting Information

Two-Photon Absorption Properties of Multipolar Triarylamino/ Tosylamido 1,1,4,4-Tetracyanobutadienes

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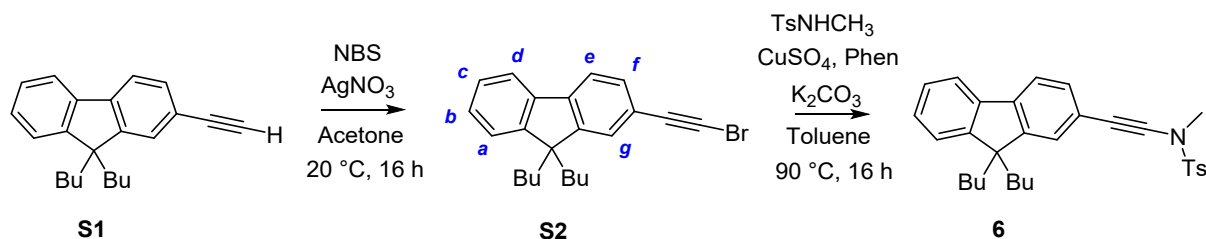
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1. Synthesis of the new alkyne precursors **6** and **7c** and of the known *tris*-TCBD derivative **5**

1.1. General

All reactions and work-up procedures of air-sensitive compounds were carried out under dry, high-purity argon or nitrogen, using standard Schlenk techniques.¹ All glassware was oven-dried overnight at 120 °C prior to use. Solvents/reagents were dried and distilled as follows: Et₂O, hexane and reagent grade THF (sodium-benzophenone), CH₂Cl₂ (CaH₂), diisopropylamine and triethylamine (KOH), and DMF (activated 3 Å molecular sieves). Flash column chromatography was performed using silica (Acros 60 Å, 40-60 mesh). Hexane used for column chromatography refers to petroleum spirit (boiling point range 60-80 °C). The required precursors **S1**² and **S3**³ were obtained as described in the literature. Other chemicals were purchased from a commercial source (Sigma-Aldrich) and used as received. Infrared spectra were obtained as KBr disks in the 400-4000 cm⁻¹ range. NMR spectra were acquired at 298 K on 400 and/or 500 MHz FT NMR spectrometers. Cyclic voltammograms were recorded in dry CH₂Cl₂ solutions (containing 0.1 M [NⁿBu₄][PF₆], purged with nitrogen and maintained under an inert atmosphere) using a Pt disk as working electrode, a Pt wire as counter electrode and an SCE reference electrode; the FeCp₂ 0/+1 couple (E_{1/2}: 0.46 V,⁴ ΔE_p = 0.09 V; I_{pa}/I_{pc} = 1) was used as an internal calibrant. High-resolution mass spectra (EI and ESI) were obtained at the Centre Regional de Mesures Physiques de l'Ouest (CRMPO, Rennes) or at Wrocław University of Science and Technology (WUST).

1.2. Synthesis of **6**:



A solution of alkyne **S1** (0.401 g, 1.33 mmol) in acetone (9 mL) was treated with N-bromosuccinimide (NBS; 0.284 g, 1.60 mmol) and AgNO₃ (23 mg, 0.13 mmol). After 16 h at 20 °C in the dark, the reaction mixture was diluted with pentane (25 mL) and the precipitate was filtered. The filtrate was concentrated under reduced pressure. The residue obtained was diluted with pentane (25 mL) and the precipitate was filtered. The filtrate was concentrated under reduced pressure to give bromoalkyne **S2** (0.468 g, 1.23 mmol, 92%) as a yellow oil. *R*_f: 0.58 (Petroleum ether); ¹H NMR (400 MHz, CDCl₃): δ = 7.71 – 7.66 (1H, m, H_g), 7.63 (1H, d, *J* = 8.2 Hz, H_e), 7.45 – 7.42 (2H, m, H_a and H_f), 7.35 – 7.30 (3H, m, H_b, H_c and H_d), 1.95 (4H, t, *J* = 8.2 Hz, CH₂-Bu), 1.14 – 1.00 (4H, m, CH₂-Bu), 0.67 (6H, t, *J* =

¹ D. F. Shriver, M. A. Drezdson, *The Manipulation of Air-Sensitive Compounds*, Wiley, New York, **1986**

² F. Malvolti, C. Rouxel, A. Triadon, G. Grelaud, N. Richy, O. Mongin, M. Blanchard-Desce, L. Toupet, F. I. Abdul Razak, R. Stranger, M. Samoc, X. Yang, G. Wang, A. Barlow, M. P. Cifuentes, M. G. Humphrey, F. Paul, *Organometallics* **2015**, *34*, 5418-5437.

³ M. L. N. Rao, D. N. Jadhav, P. Dasgupta, *Org. Lett.* **2010**, *12*, 2048-2051.

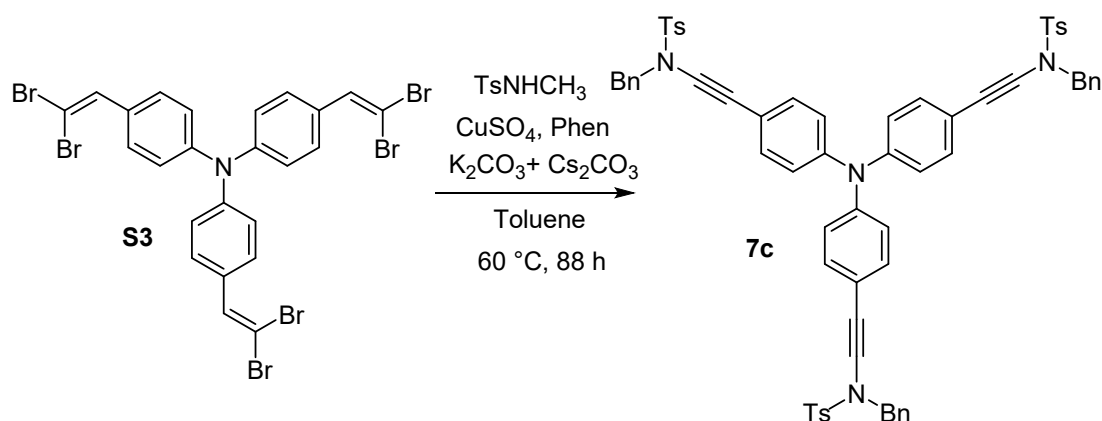
⁴ N. G. Connelly, W. E. Geiger, *Chem. Rev.* **1996**, *96*, 877-910.

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7.4 Hz, CH_{3-Bu}), 0.63 – 0.50 (4H, m, CH_{2-Bu}); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 151.0, 146.1, 142.0, 140.2, 130.9, 127.6, 126.9, 126.4, 122.9, 120.8, 120.1, 119.6, 81.1, 71.6, 55.0, 40.1, 25.9, 23.0, 13.8.

A solution of $TsNHCH_3$ (0.187 g, 1.01 mmol), $CuSO_4 \cdot 5H_2O$ (25 mg, 0.10 mmol), 1,10-phenanthroline (Phen; 36 mg, 0.20 mmol), K_2CO_3 (279 mg, 2.02 mmol) and the 2-ethynylfluorene **S2** (461 mg, 1.21 mmol) in dry toluene (2.0 mL) was heated to 90 °C under a nitrogen atmosphere for 16 h. The reaction was incomplete. $CuSO_4 \cdot 5H_2O$ (50 mg, 0.20 mmol), and 1,10-phenanthroline (73 mg, 0.40 mmol) were added and the reaction mixture was heated at 90 °C for 7 h. It was then cooled to 20 °C, diluted with Et_2O (10 mL) and filtered through Celite. The filtrate was concentrated under reduced pressure and purified by column chromatography (petroleum ether to Et_2O [4:1]) to give ynamide **6** (0.368 mg, 0.758 mmol, 75%) as a yellow solid. R_f : 0.29 (petroleum ether: Et_2O [9:1]); 1H NMR (400 MHz, $CDCl_3$): δ = 7.89 (2H, d, J = 8.3 Hz, H_{Tos}), 7.69 – 7.65 (1H, m, H_g), 7.61 (1H, d, J = 8.3 Hz, H_e), 7.42 – 7.37 (2H, m, H_{Tos}), 7.36 – 7.29 (5H, m, H_a, H_b, H_c, H_d and H_f), 3.19 (3H, s, NCH_3), 2.47 (3H, s, CH_{3-Tos}), 2.05 – 1.86 (4H, m, CH_{2-Bu}), 1.13 – 1.08 (4H, m, CH_{2-Bu}), 0.68 (6H, t, J 7.4 Hz, CH_{3-Bu}), 0.64 – 0.50 (4H, m, CH_{2-Bu}); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ = 151.1, 150.9, 145.0, 141.3, 140.6, 133.5, 130.7, 130.0, 128.1, 127.6, 127.0, 126.1, 123.1, 121.0, 120.1, 119.7, 77.4, 70.2, 55.2, 40.3, 39.6, 26.1, 23.2, 21.9, 14.0; HRMS (ESI): calculated for $C_{31}H_{35}NNaO_2S$ $[M+Na]^+$ 508.22862, found 508.228.

1.3. Synthesis of 7c

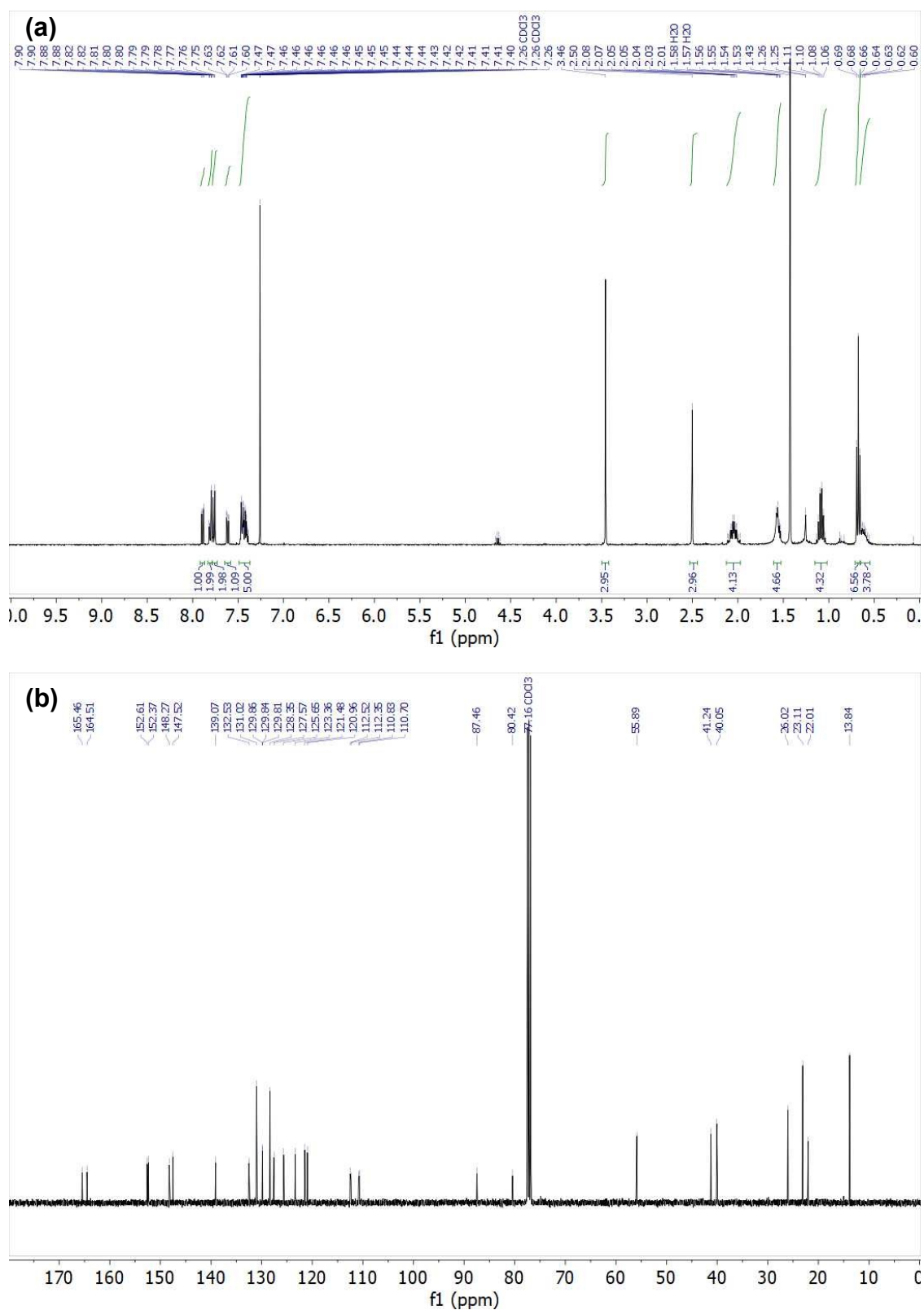


A solution of dibromoalkene **S3** (131 mg, 0.166 mmol), $TsNHBn$ (125 mg, 0.478 mmol), $CuSO_4 \cdot 5H_2O$ (13 mg, 0.053 mmol), 1,10-phenanthroline (38 mg, 0.211 mmol), K_2CO_3 (189 mg, 1.367 mmol) and Cs_2CO_3 (373 mg, 1.145 mmol) in dry deoxygenated toluene (2 mL) was heated to 60 °C under a nitrogen atmosphere over 88 h. The reaction mixture was cooled to room temperature, diluted with Et_2O (10 mL) and filtered through Celite. The filtrate was concentrated under reduced pressure and purified by column chromatography (cyclohexane/ CH_2Cl_2 [1:0] to [0:1]) to give ynamide **7c** (68.7 mg, 0.063 mmol, 38%) as a brown solid. R_f : 0.50 (cyclohexane/ $EtOAc$ [7:3]). 1H NMR (400 MHz, $CDCl_3$): δ = 7.81 (d, J = 7.9 Hz, 6H), 7.37 – 7.28 (m, 21H), 7.12 (d, J = 8.2 Hz, 6H), 6.90 (d, J = 8.3 Hz, 6H), 4.58 (s, 6H), 2.46 (s, 9H). ^{13}C NMR (101 MHz, $CDCl_3$): δ = 146.3, 144.7, 134.7, 134.5, 132.6, 129.7, 128.9, 128.5, 128.3, 127.8, 123.8, 117.3, 82.4, 71.0, 55.8, 21.7. HRMS (ESI, CH_2Cl_2): calculated for $C_{66}H_{54}N_4O_6S_3$ $[M]^+$ 1094.3205, found 1094.3198.

1.4. Synthesis of **5**

In a round bottom flask, 20 mL of CH₂Cl₂ was added to tris(4-(ferrocenylethynyl)phenyl)amine (**9**; 87 mg, 0.10 mmol) and tetracyanoethene (TCNE) in slight excess (45 mg, 0.35 mmol). The resulting mixture was stirred for 24 h at 20 °C. The solvent was removed under reduced pressure. The resulting solid was absorbed on silica and loaded at the top of a chromatography column (silica gel, 3 x 15 cm) and eluted with CH₂Cl₂, to give the known⁵ product **5** as a dark green solid (93 mg; 75 %). Black crystals of this compound were obtained by slow diffusion of toluene into a saturated solution of **5** in CH₂Cl₂. *R_f*: (CH₂Cl₂) = 0.3. ¹H-NMR (400 MHz, CDCl₃): δ = 7.56 (s, 6H), 7.13 (s, 6H), 5.49 (s, 3H), 5.07 (s, 3H), 4.90 (s, 3H), 4.47 (s, 18H). ¹³C{¹H}-NMR (101 MHz, CDCl₃): δ = 172.6, 164.3, 149.7, 131.2, 127.5, 125.1, 113.6, 113.0, 112.1, 111.9, 84.6, 78.9, 76.4, 75.4, 75.1, 73.0, 73.0, 71.3. IR (KBr, cm⁻¹): $\bar{\nu}$ = 2222 (m, C≡N), 1590 (s, C=CAr), 1522 (s, C=CAr), 1499 (m, C=CAr). HRMS (ESI): Calcd.: 1276.1397 [M+Na]⁺; found: 1276.1377 [M+Na]⁺. The spectroscopic data correspond to those previously reported in the literature for this compound.

⁵ T. Shoji, N. Kamata, A. Maruyama, S. Ito, T. Okujima, *Bull. Chem. Soc. Jpn.* **2015**, *88*, 1338-1346.

2. ^1H / $^{13}\text{C}\{^1\text{H}\}$ NMR spectra of selected compounds

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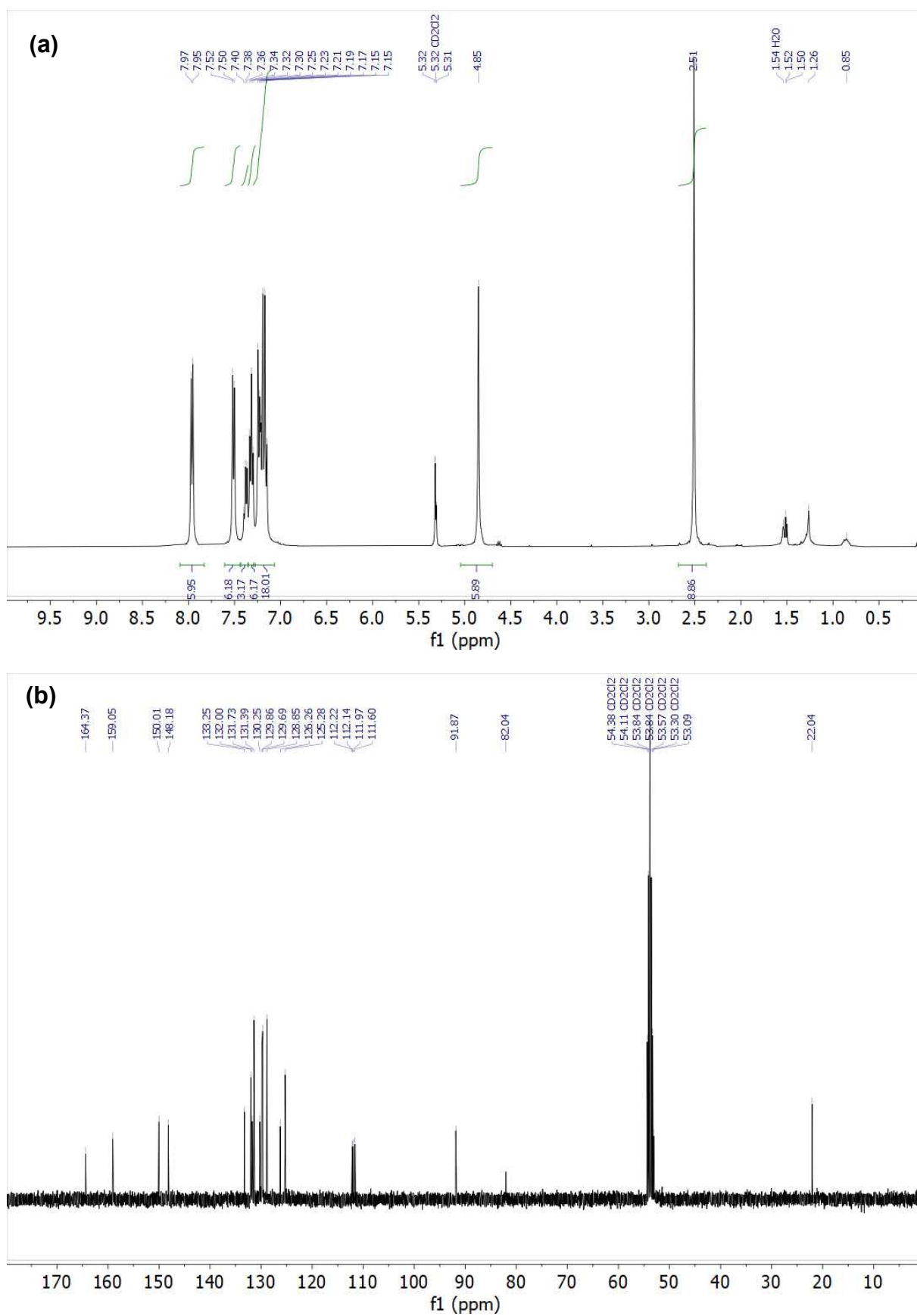


Figure S2. ^1H (a) and $^{13}\text{C}\{^1\text{H}\}$ (b) NMR spectra at 400 and 100 MHz, respectively, for **3c** in CD_2Cl_2 .

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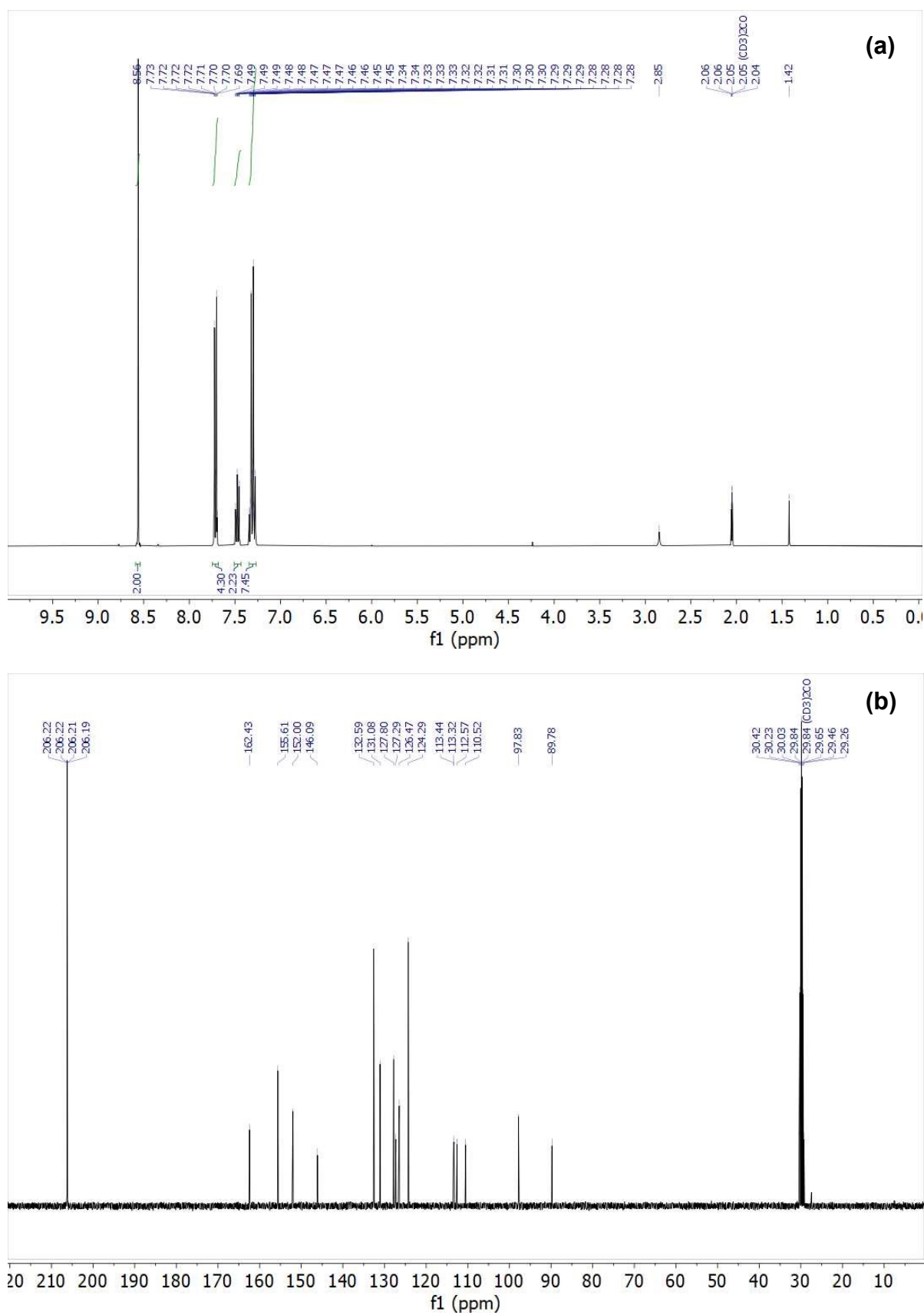


Figure S3. ^1H (a) and $^{13}\text{C}\{^1\text{H}\}$ (b) NMR spectra at 400 and 100 MHz, respectively, for **4b** in acetone- d^6 .

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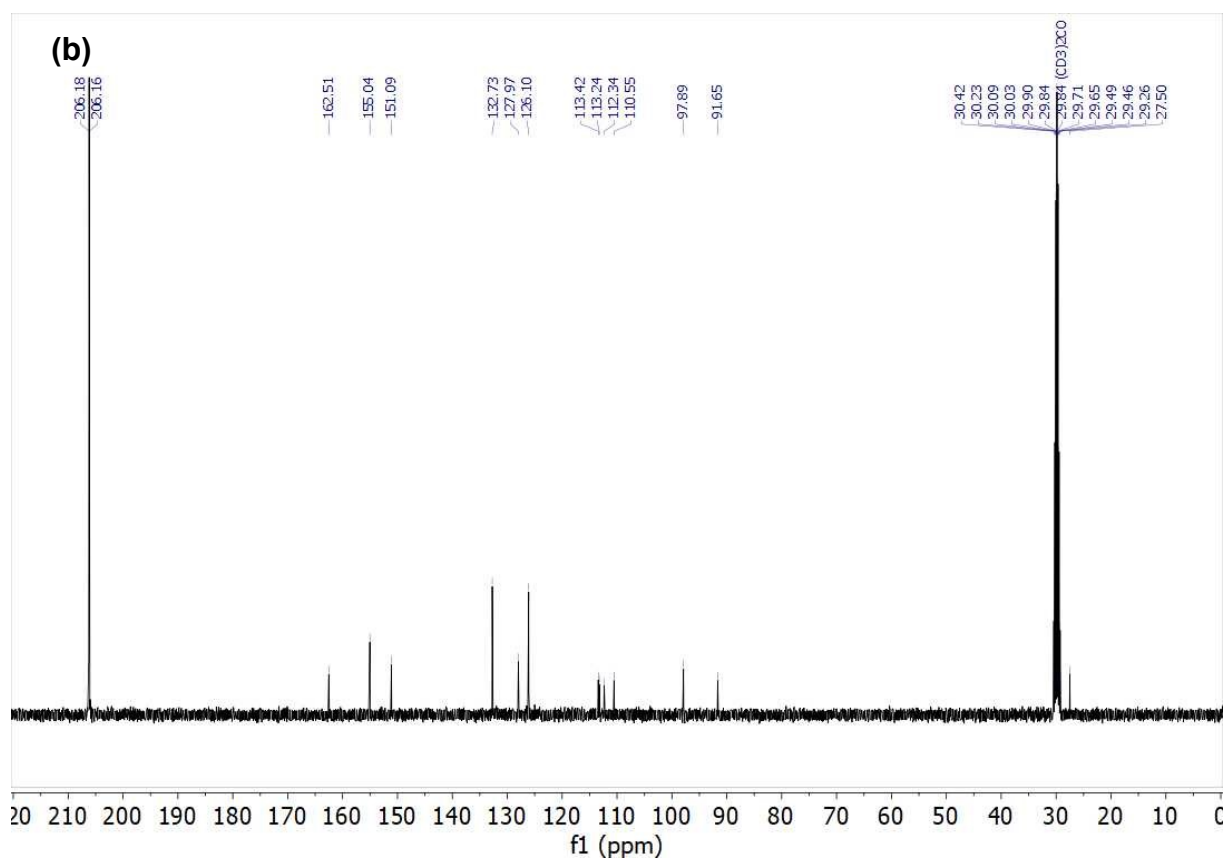
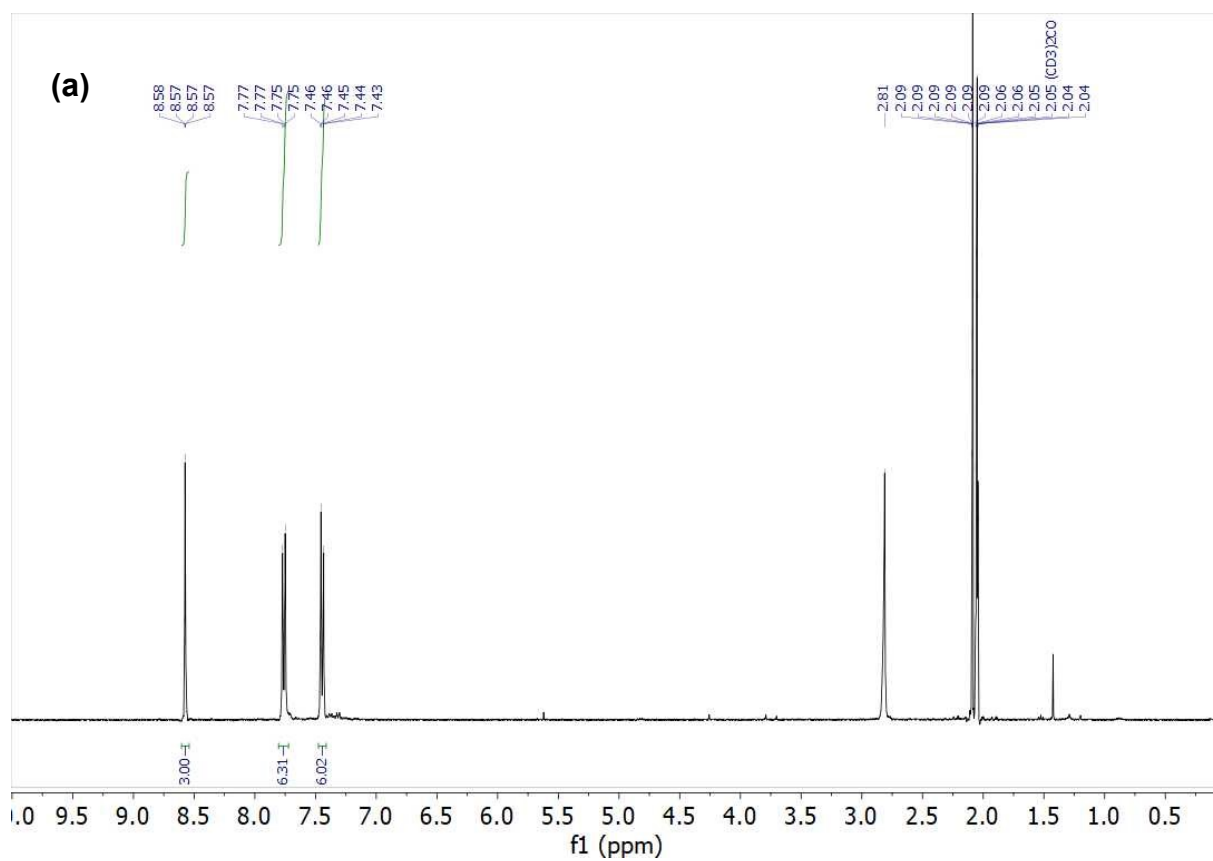


Figure S4. ^1H (a) and $^{13}\text{C}\{^1\text{H}\}$ (b) NMR spectra at 400 and 100 MHz, respectively, for **4c** in in acetone- d^6 .

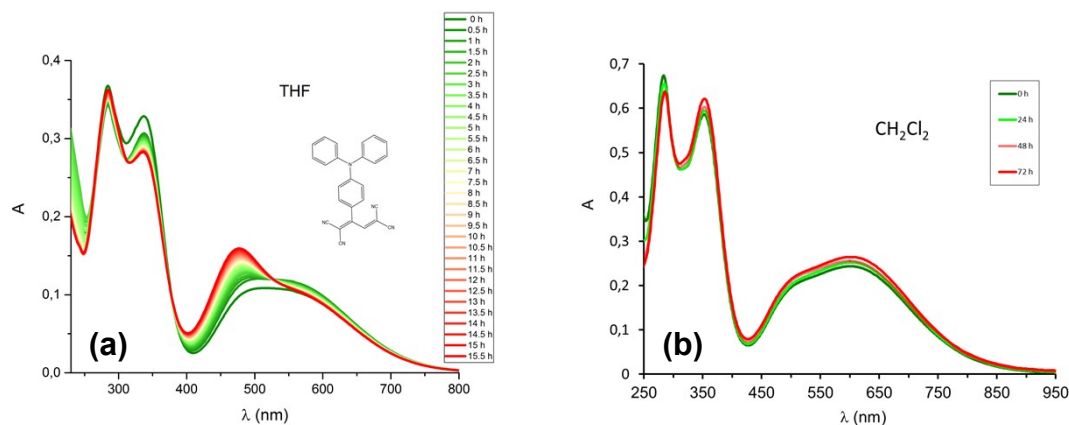
3. Evolution of compound **4a** in THF solution

Figure S5. UV-Vis spectra of compound **4a** dissolved in THF (a) and CH_2Cl_2 (b) at 20 °C.

Evolution of **4a in THF solution.** As revealed by UV-visible spectroscopic monitoring, and in contrast to **4b-c**, the compound **4a** slowly evolves over time when left in THF at 20 °C (Figure S5a), forming several (unknown) products (TLC evidence), but does not transform appreciably when CH_2Cl_2 (Fig. S5b) or CDCl_3 are used as solvent (^1H NMR evidence). We have verified that this process is apparently not influenced by water (H_2O), since adding some drops to the THF solution slows the process slightly. The transformation also occurs in the dark.

ESI-MS monitoring. Consistent with the literature, when dissolved in acetonitrile and subjected to ESI-MS (positive mode), **4a** gives a positive ion at 397.1327 corresponding to the molecular ion $[\text{M}]^+$ (calc 397.1328).⁶ In an attempt to identify the product(s) formed, we have monitored aliquots of the THF solution over time (0, 5, 16 and 24 h) in methanol (containing 1% formic acid) by ESI-MS. In positive mode, the molecular cation corresponding to protonated **4a** at 398.1428 amu ($[\text{M} + \text{H}]^+$, calc 398.1406 amu) is weakly observed in the first spectrum, along with a new cation at 430.1656 amu ($[\text{M} + \text{H} + \text{MeOH}]^+$, calc 430.1668 amu) which rapidly develops and dominates the spectra after 16 h, while the cation at 398.1428 amu has totally disappeared at this time. When monitored in negative mode, these mixtures revealed the initial presence of an anion at 396.1254 am ($[\text{M} - \text{H}]^-$, calc 396.1249 amu) corresponding to the deprotonated compound **4a**, which slowly disappeared with concomitant appearance of anions at 412.1177 amu ($[\text{M} + \text{O} - \text{H}]^-$, calc 412.1198 amu) and 793.3129 amu ($[2\text{M} - \text{H}]^-$, calc 793.2576 amu), the first-mentioned of these anions becoming dominant over the second in the spectra after 16 h while the anion at 396.1254 amu is still weakly present.

This reaction seems to be retarded by adding acid (one drop of HCl in water) to the medium (however the anion $[\text{M} - \text{H}]^-$ can still be detected at 396.1254 amu by ESI-MS (negative mode) in this case), while base (NEt_3) accelerates the disappearance of **4a** and leads to an immediate color change of the medium from purple to orange (new cations at 373.2420 amu ($[\text{M} + 2\text{H} - \text{CN}]^+$, calc 373.1453 amu) and 598.3777 amu ($[\text{M} + 2\text{NEt}_3 - \text{H}]^+$, calc 598.3658 amu) are observed by ESI-MS (positive mode) in this case). In accordance with these observations, we tentatively propose a mechanism for

⁶ X. Tang, W. Liu, J. Wu, C.-S. Lee, J. You, P. Wang, *J. Org. Chem.* **2010**, *75*, 7273-7278.

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the decomposition of **4a** in THF (Figure S6). The evolution of the compound in THF would be driven by the formation of **4a'** as an intermediate, leading to the blue shift of the $\text{NPh}_2 \rightarrow \text{C}(\text{CN})_2$ CT absorption at lowest energy.

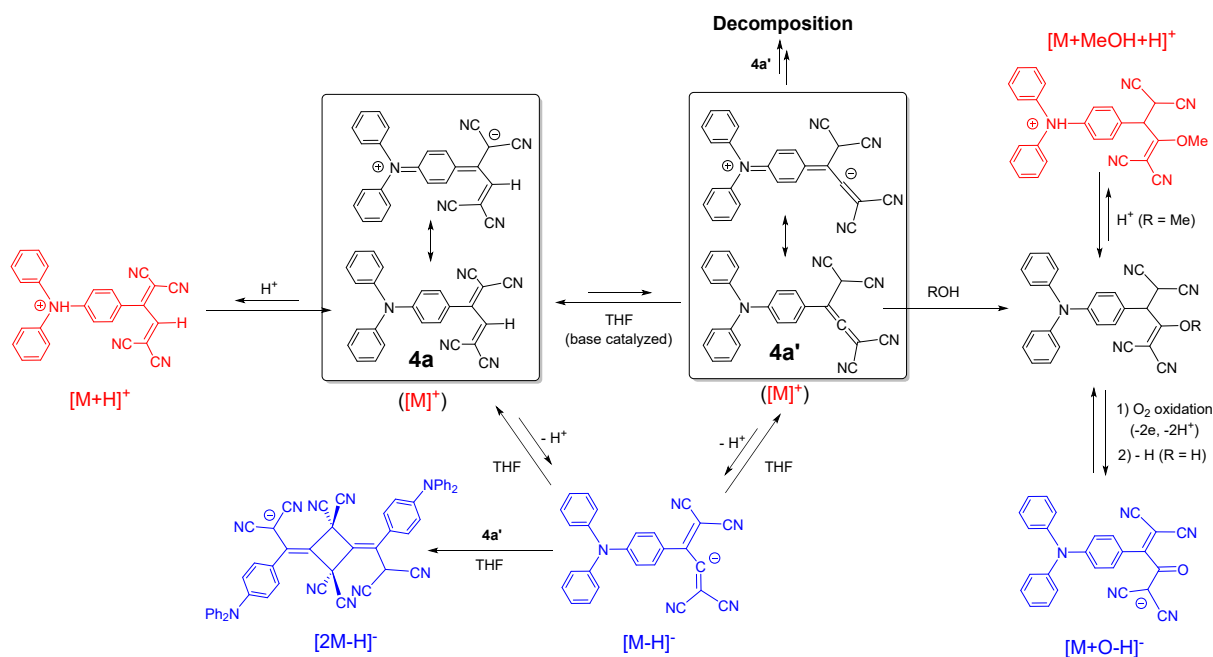


Figure S6. Tentative mechanism for the decomposition of **4a** in THF at 20 °C, based on MS monitoring. The positive and negative ions that have been detected by ESI-MS are indicated in red and blue, respectively.

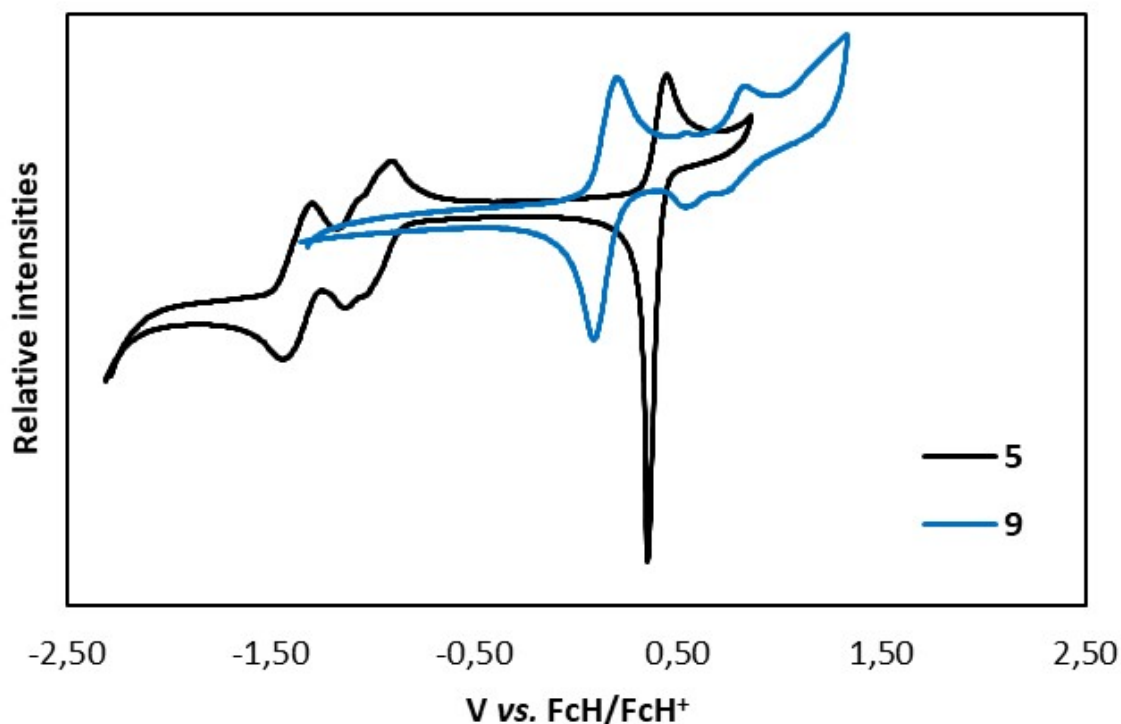
3. Cyclic voltammograms of **5** and **9** and derivation of ΔG_{CT} values

Figure S7. Cyclic voltammograms of **5** and **9** in $\text{CH}_2\text{Cl}_2/[n\text{-Bu}_4\text{N}][\text{PF}_6]$ (0.1 M) at 25 °C at 0.1 V/s.

Derivation of ΔG_{CS} values (eV)

ΔG_{CS} values were obtained using eq 1 as described previously.⁷ Estimates of the donor-acceptor distances „ d ” in eq. 1 for **3a**, **4a** and **5** were obtained from the solid state structures of **3c**, **4b** and **5**, considering the distances between the donor group (nitrogen of Fe atom) and the centroid of the TCBD moieties *i.e.* 6.0, 6.0 and 3.5 Å, respectively. The ΔG_{CS} value for **A_H** was taken from the previous work.⁷

$$\Delta G_{CS} = E^\circ(\text{D}^+/\text{D}) - E^\circ(\text{A}/\text{A}^-) - e^2/4\pi\epsilon_0\epsilon d \quad (1)$$

The CV data measured in CH_3CN for **3a** were corrected for difference in solvation energies and the corresponding ΔG_{CS} value was estimated in CH_2Cl_2 .

⁷ Z. Pokladek, N. Ripoche, M. Betou, Y. Trolez, O. Mongin, J. Olesiak-Banska, K. Matczyszyn, M. Samoc, M. G. Humphrey, M. Blanchard-Desce, F. Paul, *Chem. Eur. J.* **2016**, 22, 10155-10167.

4. X-ray data for 1, 3c, 4b, 5 and 9

Crystallization of tris(4-(2-ferrocenylethynyl)phenyl)amine (9). Crystals of the title compound were grown by slow diffusion of *n*-hexane into a solution of 9 in CH₂Cl₂.

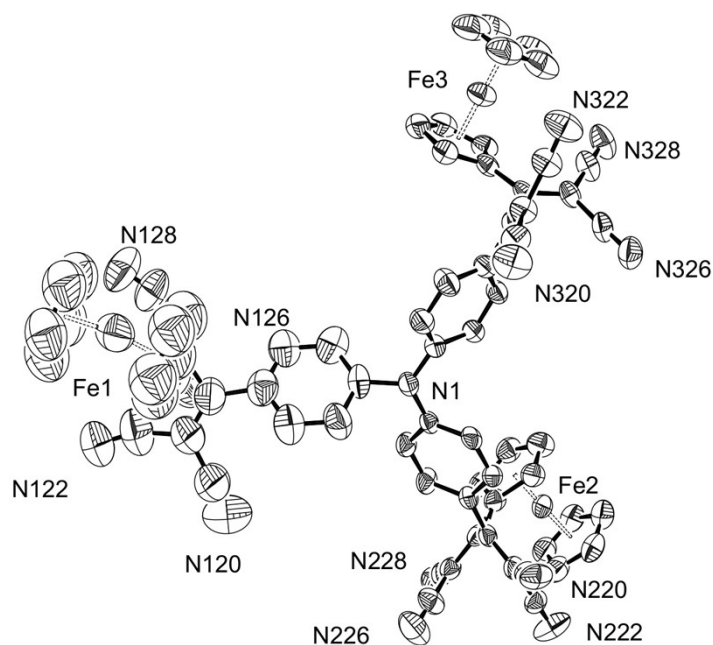


Figure S8. ORTEP representation of **5** at the 50 % probability level. Hydrogen atoms have been omitted.

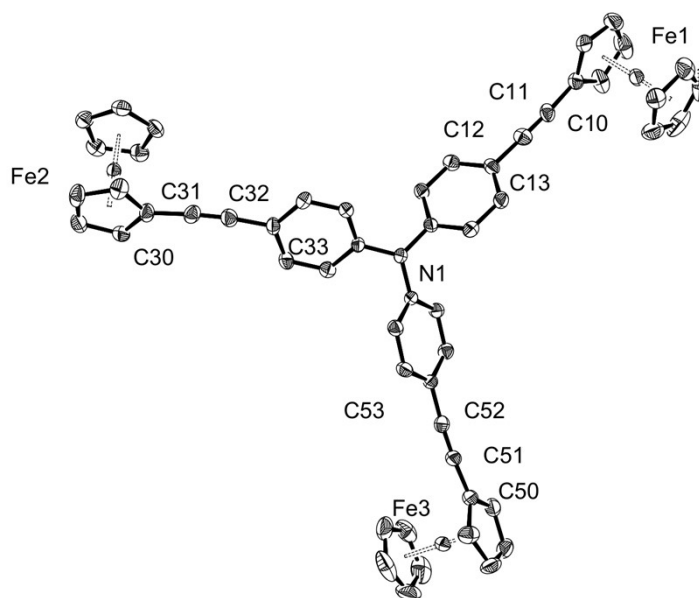


Figure S9. ORTEP representation of **9** at the 50 % probability level (CCDC 2098561). Hydrogen atoms have been omitted. Selected bond lengths (Å); C11-C12, 1.161(5); C31-C32, 1.183(5); C51-C52, 1.186(5).

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Table S1. Crystal Data, Data Collection, and Refinement Parameters for **1**, **3c**, **4b**, **5** and **9**.

Cmpd	1	3c	4b	5•C ₇ H ₈	9•CH ₂ Cl ₂
formula	C ₇₇ H ₇₆ N ₁₀ O ₄ S ₂	C ₈₄ H ₅₄ N ₁₆ O ₆ S ₃	C ₃₄ H ₁₅ N ₉	C ₇₂ H ₃₉ Fe ₃ N ₁₃ , C ₇ H ₈	C ₅₄ H ₃₉ Fe ₃ , CH ₂ Cl ₂
fw	1269.59	1479.61	549.55	1345.85	954.34
cryst. syst.	triclinic	trigonal	monoclinic	monoclinic	monoclinic
space group	<i>P</i> -1	<i>R</i> -3	<i>P</i> 2/ <i>a</i>	<i>P</i> 2 ₁ / <i>c</i>	<i>P</i> 2 ₁ / <i>c</i>
<i>a</i> (Å)	12.2334(10)	21.023(2)	10.7358(5)	19.293(4)	13.9363(3)
<i>b</i> (Å)	16.0882(15)	21.023	9.7979(4)	16.414(3)	20.8729(4)
<i>c</i> (Å)	18.7919(16)	30.595(3)	13.1395(4)	23.333(5)	15.9589(3)
α (deg)	107.984(3)	90.0	90.0	90.0	90.0
β (deg)	97.849(3)	90.0	93.928(3)	93.60(3)	110.8000(10)
γ (deg)	90.566(3)	120.0	90.0	90.0	90.0
<i>V</i> (Å ³)	3479.5(5)	11710(3)	1378.87(10)	7374(3)	4339.74(15)
Z	2	6	2	4	4
<i>D</i> (calcd) (g cm ⁻³)	1.212	1.259	1.324	1.212	1.461
crystal size (mm)	0.37 × 0.35 × 0.15	0.22 × 0.10 × 0.06	0.26 × 0.22 × 0.02	0.08 × 0.06 × 0.05	0.27 × 0.18 × 0.08
<i>F</i> (000)	1344	4596	564	2760	1960
abs. coef. (mm ⁻¹)	0.134	0.159	0.084	0.633	1.152
N° total refl. / N° unique refl.	80195/15904	13829/5970	11230/3589	34351/9824	39978/9935
N° of variables / restraints / N° refl. > 2σ(<i>I</i>)	846/20/11894	329/0/4152	196/0/3589	814/0/6046	550/0/6515
<i>R</i> _{int}	0.0593	0.0460	0.0450	0.0986	0.0562
final <i>R</i>	0.0539	0.0611	0.0474	0.1342	0.0501
<i>R</i> _w	0.1269	0.1460	0.1149	0.4105	0.1207
Largest diff. peak and hole (e.Å ⁻³)	1.155 / -0.771	0.489 / -0.444	0.242 / -0.210	- / -	0.710 / -0.754
Goodness of fit / <i>F</i> ² (<i>S</i> _w)	1.009	1.008	1.009	1.497	1.035

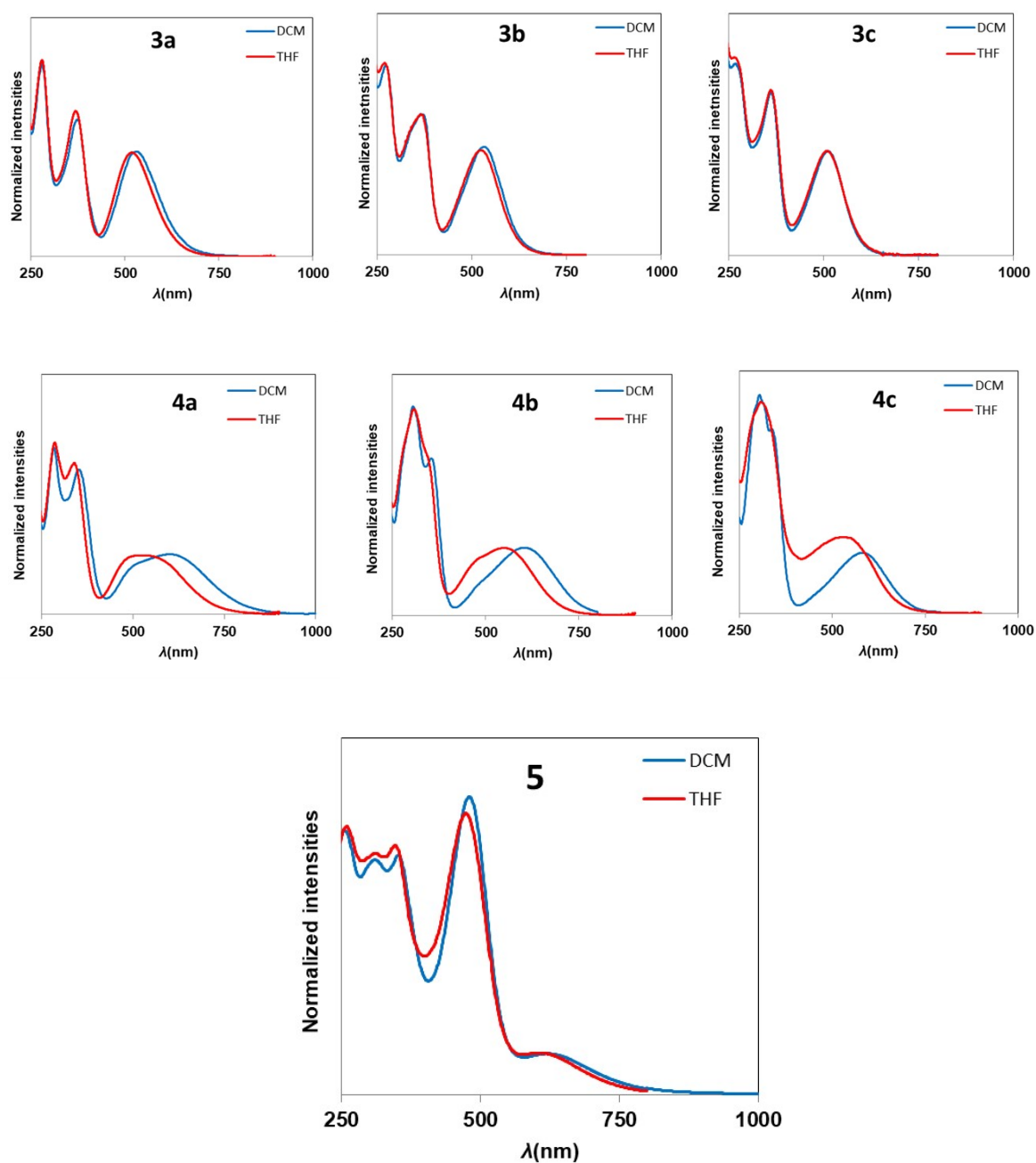
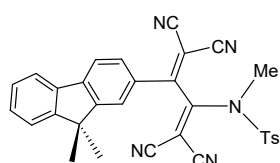
5. Solvatochromism in CH₂Cl₂ and THF for selected compounds

Figure S10. Solvatochromism of compound **3a-c**, **4a-c** and **5** in CH₂Cl₂ and THF at 20 °C. For **4a**, the time-dependent spectral evolution indicates that this compound undergoes a slow decomposition in THF (see Figure S5).⁸

⁸ **UV-Vis** (THF) of **4a**: λ_{max} (nm) [ϵ ($10^3 \text{ M}^{-1} \cdot \text{cm}^{-1}$)] = 552 [8.4], 490 [sh, 7.3], 308 [25.7], 290 [sh, 22.9]. Corresponding data for **4b-c** given in experimental part.

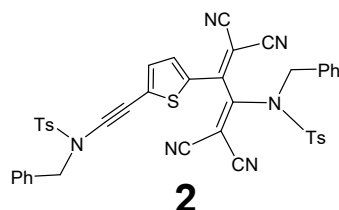
7. Cartesian coordinates of the DFT optimized geometries of 1'-6



1'

S	-2.80801000	0.35197900	-1.06126500
O	-3.11499200	1.68521400	-1.90728500
O	-1.60096900	-0.63948800	-1.46618100
N	-2.13027800	4.06755400	0.45768100
N	1.33326500	3.52649300	-2.10655400
N	-2.68265800	-1.12851800	3.71218800
N	1.44782000	0.42980000	3.57322900
N	-2.43404800	0.78881600	0.68335700
C	5.76015500	-0.65314100	-0.36393700
C	7.12634600	-0.90484200	-0.48500500
H	7.84793000	-0.09420900	-0.47178500
C	7.56492400	-2.23321900	-0.62530100
H	8.62534600	-2.44292700	-0.71977100
C	6.64360900	-3.29629700	-0.64426100
H	7.00198200	-4.31438600	-0.75331000
C	5.26929300	-3.04811100	-0.52339500
H	4.56175200	-3.87064300	-0.53887300
C	4.83519300	-1.72293800	-0.38421700
C	3.48749700	-1.16871200	-0.24277500
C	2.23384300	-1.79430300	-0.18819000
H	2.14273300	-2.87154100	-0.27197300
C	1.09115500	-1.01054200	-0.03038700
H	0.11429800	-1.47713400	-0.02167800
C	1.18161600	0.40019400	0.06108900
C	2.45464600	1.01939600	0.00729200
H	2.54204500	2.09319900	0.12753500
C	3.59040100	0.23975600	-0.14094400
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H	6.53923000	1.64050500	1.07787100
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C	5.30235000	1.61470800	-1.41261000
H	4.72861400	2.54203600	-1.31691100
H	6.36353100	1.87782100	-1.47427700
C	-0.03461600	1.19804800	0.24076500
C	-0.22601100	2.45276600	-0.31075300
C	-1.29649800	3.31564200	0.09796800
C	0.64970400	3.02259100	-1.28898000
C	-1.12363800	0.67124600	1.11653700
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C	-1.85697200	-0.52866200	3.12076900
C	0.42102300	0.30856200	3.00672100
C	-3.57084800	1.25836200	1.48910700
H	-4.20740700	0.44294500	1.83846600
H	-4.15040800	1.97164500	0.89504700
H	-3.17761700	1.80407100	2.34840800
C	-4.38037600	-0.59031100	-0.88351200
C	-5.56260600	0.03963800	-1.26527400

H	-5.54717700	1.05382500	-1.64970500
C	-6.75268800	-0.69143800	-1.15797200
H	-7.68775500	-0.22704100	-1.45150800
C	-6.75367100	-2.01584600	-0.68166700
C	-5.52594400	-2.61008900	-0.31950800
H	-5.51273300	-3.63349600	0.04053200
C	-4.32364300	-1.90543000	-0.41923600
H	-3.37763200	-2.36753200	-0.16020700
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H	-8.89401600	-2.22469400	-0.92422000
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H	-7.98580100	-3.72796500	-1.13619000
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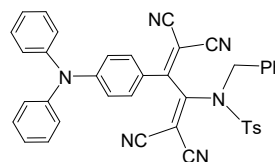


2

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C	4.73939100	-1.99670300	2.12798600
N	5.87500900	-1.98514500	1.80894800
C	2.74921800	-1.51682700	0.19910300
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C	2.84257300	-3.96593600	0.24554500
N	2.63462900	-5.01825200	0.73493200
C	3.72079400	-2.82666100	-1.68382300
N	4.28376600	-2.95589200	-2.71259400
N	2.68645400	-0.31772200	-0.47391400
C	2.46619300	-0.12881100	-1.93978100
H	3.23554400	-0.65922400	-2.50234500
H	2.61018600	0.93827100	-2.13014600
C	1.07823700	-0.52653000	-2.39422200
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H	0.11947800	1.13299900	-1.40363200
C	-1.31100600	-0.07605100	-2.48317300
H	-2.16198800	0.53085500	-2.19408100
C	-1.50037800	-1.21031900	-3.28876800
H	-2.49792800	-1.47933800	-3.61902700
C	-0.39841000	-1.99428200	-3.65588800
H	-0.53633600	-2.86900500	-4.28233400
C	0.88800900	-1.65314600	-3.21129200
H	1.73716100	-2.25868500	-3.51085300
S	3.07289900	1.29104800	0.37768500
O	3.24824100	0.99171400	1.94430600
O	1.94999200	2.32480800	-0.14555100

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C	5.90321300	3.21990200	-1.84626600
H	5.92632800	4.03466600	-2.56173700
C	7.10203200	2.57603100	-1.48628900
C	7.05407400	1.52836500	-0.54246600
H	7.96974600	1.02613700	-0.24916900
C	5.84679000	1.11873300	0.02902100
H	5.82899300	0.31993200	0.76182400
C	8.41883200	2.99027200	-2.09192200
H	8.30887500	3.86006900	-2.74323300
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H	9.14787500	3.23906000	-1.31401500
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C	0.53615600	-0.84434600	3.27146300
S	-0.32271300	-1.45655100	0.85579100
C	-0.86165000	-0.65008000	3.30581600
H	1.17781000	-0.63213900	4.11411900
C	-1.50594900	-0.92010500	2.10147700
H	-1.40132200	-0.30502700	4.17694400
C	-2.85427800	-0.72844300	1.78196600
C	-4.00698600	-0.49780500	1.42296500
N	-5.22470800	-0.12922900	1.05381800
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H	-7.22380000	-0.46377900	0.58966200
H	-6.38015000	-1.85954200	1.29208700
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C	-6.35374500	-0.78210000	-1.94312400
C	-5.45648800	-2.83264800	-1.00587500
C	-6.11533400	-1.26831600	-3.23498400
H	-6.80457400	0.19523900	-1.79598500
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H	-5.20425300	-3.44238900	-0.14296900
C	-5.54703000	-2.54014800	-3.41578800
H	-6.38327900	-0.66667000	-4.09746600
H	-4.78194000	-4.30601800	-2.43283300
H	-5.37448300	-2.92178100	-4.41701300
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O	-6.94566100	1.86105500	0.32613700
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C	-4.04011300	2.77637000	-0.58285100
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H	-4.79599000	2.47182000	-1.29828900
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C	-2.12978500	3.59331300	1.31399100
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H	-1.39127100	3.90782600	2.04378500
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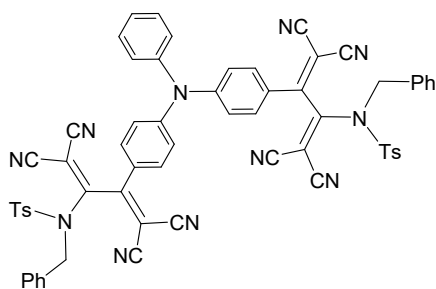


3a

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C	1.02703400	0.79279300	0.72629200
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C	0.37643400	-0.32001400	1.30900700
C	1.09998000	-1.53243500	1.40403000
H	0.61756500	-2.41977200	1.79343900
C	2.41291800	-1.62237200	0.96390700
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S	-2.43776500	-1.20861000	-0.88934000
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C	-4.26010500	-1.33510900	-1.15710200
C	-4.72029100	-1.30305400	-2.47260300
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Supporting Information

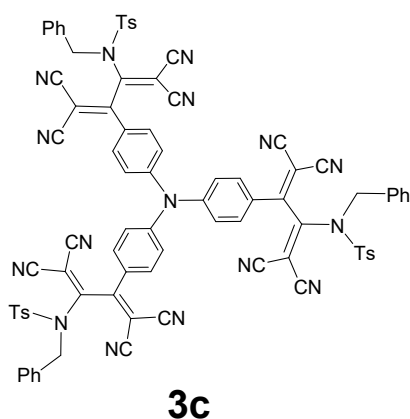
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H	4.25334200	-4.47027800	-2.50858500	H	-2.76849100	-0.28561700	-2.30632300
C	6.06416900	-4.26227100	-1.34612800	C	-2.80001900	1.27950700	-3.80089400
C	6.76820100	-3.44856500	-0.44552800	H	-1.72101500	1.39186100	-3.82348100
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C	6.21648000	-2.24100500	-0.00057800	H	-3.15097300	2.80725600	-5.29137200
H	6.75548900	-1.61685600	0.70333800	C	-4.99826100	1.92021900	-4.60792700
C	5.24608300	0.57627800	-0.04991500	H	-5.62547900	2.52944700	-5.24957000
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H	6.17651400	3.21917900	1.89589700	C	-5.21182900	-4.39056700	-0.66724600
C	5.30336800	1.43869100	1.06068400	H	-4.32862200	-4.34551400	-1.29485600
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H	-2.01806200	3.38912900	-1.26045500	C	4.88075900	0.48749800	1.81163100
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N	-4.93594800	2.25199600	4.64553100	N	5.72555200	-4.40801000	1.65845300
C	-6.93326200	0.23170900	2.68607700	N	5.11407100	-1.11086900	-0.08790600
N	-7.87711800	-0.45265100	2.86258000	C	4.61519200	-2.29973900	-0.84143100
C	-5.80877600	0.46042400	0.09687900	H	5.09579900	-3.20521800	-0.46906100
C	-6.97933200	1.08872100	-0.30973100	H	4.94176900	-2.16230200	-1.87570000
C	-7.34815300	2.36785900	0.21552700	C	3.10682700	-2.41857000	-0.80642300
N	-7.67752700	3.43294800	0.59944500	C	2.31815800	-1.55148500	-1.58515900
C	-7.89738800	0.53703400	-1.25050500	H	2.80408600	-0.79119800	-2.18940100
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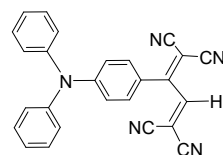
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H	3.08070000	-4.09691700	0.55915400	C	-4.24711600	-6.78024800	2.58292000
S	5.97691400	0.11125100	-1.18776300	N	-4.73975300	-7.82387400	2.34123500
O	6.31265500	1.40628600	-0.29915800	C	-4.12709400	-4.76205700	0.60328200
O	5.05652800	0.21889500	-2.50635200	C	-5.51441600	-4.71624700	0.66047200
C	7.52928700	-0.77529400	-1.64793300	C	-6.19285700	-4.27528300	1.84080300
C	7.61680200	-1.29157800	-2.93986600	N	-6.79501100	-3.89444500	2.78017800
H	6.80154400	-1.16288300	-3.64332900	C	-6.36775300	-5.09502700	-0.41699300
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H	8.89804900	-2.35821900	-4.29670700	N	-3.43811900	-4.86363200	-0.58598700
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C	8.55709000	-0.86852300	-0.70722000	C	-3.98552500	-2.85815300	-1.97180300
H	8.47050100	-0.43866200	0.28432000	C	-2.81317400	-2.08490100	-2.05682200
C	11.12915900	-2.80794700	-2.76634600	H	-1.84963500	-2.58561000	-2.07028300
H	11.13642000	-3.07764300	-3.82476600	C	-2.89245400	-0.68833100	-2.10774200
H	11.24098000	-3.72933500	-2.18417100	H	-1.98424900	-0.09808300	-2.17625900
H	12.01055700	-2.19015100	-2.56716300	C	-4.14068400	-0.04427200	-2.06635600
C	0.18885900	4.49507800	-0.34740900	H	-4.19474300	1.03928200	-2.07842200
C	0.99276100	4.58260200	-1.49723200	C	-5.31066100	-0.81445600	-1.99811000
H	1.54884900	3.71378700	-1.83235800	H	-6.27953700	-0.32735900	-1.97464300
C	1.07743700	5.79190200	-2.19840800	C	-5.23526800	-2.21556400	-1.95715300
H	1.70327700	5.85538000	-3.08202400	H	-6.14830600	-2.80161700	-1.92622600
C	0.35082600	6.91292000	-1.76817800	S	-1.85279200	-5.81846200	-0.67637000
C	-0.45697000	6.81949100	-0.62455900	O	-1.39350300	-6.12577700	0.83097100
H	-1.01642000	7.68284800	-0.28135000	O	-0.90963900	-4.97027200	-1.67384500
C	-0.53412600	5.61757900	0.09003800	C	-2.33603300	-7.37733000	-1.53684500
H	-1.14609800	5.54606300	0.98243100	C	-1.99067000	-7.50941400	-2.88103600
H	0.41533000	7.84718200	-2.31495900	H	-1.44512000	-6.72494300	-3.39364100



N	-0.00007800	0.00079700	2.30868200	C	-0.52204700	-2.32284100	2.92410500
C	-0.86124000	-1.13109100	2.25042800	H	0.38084300	-2.36703700	3.52107700
C	-2.04371500	-1.09338700	1.48483600	C	-3.36990900	-4.60137900	1.89442900
H	-2.32157900	-0.19284900	0.95222800	C	-3.58060200	-5.54564500	2.87461600
C	-2.85171100	-2.22124400	1.38397600	C	-3.10546300	-5.42385600	4.21770000
H	-3.75341200	-2.16176700	0.78495200	N	-2.75335500	-5.35950000	5.34067900
C	-2.51230500	-3.42317000	2.04144600	C	-4.24711600	-6.78024800	2.58292000
C	-1.32735000	-3.45105200	2.81120600	N	-4.73975300	-7.82387400	2.34123500
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Supporting Information

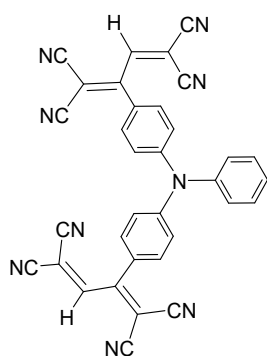
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C	6.79491600	-3.22588100	1.83753700	C	-0.48433200	4.87579600	-1.97432300
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C	7.59329800	-2.96915300	-0.42007500	H	-1.31892700	2.89116700	-2.07801600
N	8.25802100	-3.43198100	-1.27786900	C	0.84555100	2.84260600	-2.11186500
N	5.93215800	-0.54428700	-0.58932700	H	0.90123000	1.76102100	-2.18238200
C	5.72716000	-1.18001700	-1.92618300	C	2.02845200	3.59979200	-2.06725200
H	6.60760800	-1.76052100	-2.20469900	H	2.99314800	3.10340500	-2.07890800
H	5.63911000	-0.35683800	-2.64014900	C	1.94838800	4.99811500	-1.99668300
C	4.46865700	-2.01801700	-1.97769400	H	2.85550500	5.59225600	-1.97079000
C	3.21386500	-1.38703800	-2.06087700	C	0.69814400	5.63509300	-1.95658900
H	3.16765900	-0.30209500	-2.07141500	H	0.64865300	6.71885100	-1.92379900
C	2.04286300	-2.15201000	-2.11358200	S	-4.11384100	4.51227500	-0.67798500
H	1.07831500	-1.65896400	-2.18043500	O	-4.60819000	4.26820900	0.82982500
C	2.10697800	-3.55528900	-2.07605200	O	-3.85174500	3.27148100	-1.67576000
H	1.19457600	-4.14223500	-2.08948800	C	-5.22328000	5.70991300	-1.53760300
C	3.35791800	-4.18562300	-2.00966000	C	-5.51337000	5.47507100	-2.88085100
H	3.41882500	-5.26839700	-1.98900900	H	-5.10882600	4.60908800	-3.39289400
C	4.53484800	-3.42172200	-1.96674200	C	-6.36536100	6.37916500	-3.52740500
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S	5.96864700	1.30614800	-0.67881000	C	-6.90816200	7.48463000	-2.84562200
O	6.00049800	1.85682500	0.82889600	C	-6.58911500	7.66716400	-1.48335300
O	4.76636500	1.70065900	-1.68025200	H	-7.00475200	8.51009300	-0.94173600
C	7.56355900	1.66571400	-1.53387000	C	-5.74422600	6.78189800	-0.80958100
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H	6.55903300	2.12017000	-3.39167000	C	-7.81603000	8.46158000	-3.54811900
C	8.72099000	2.31960200	-3.51990700	H	-8.02396400	8.15416700	-4.57527300
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H	10.87789400	1.80201900	-0.92851100				
C	8.75020500	1.57827400	-0.80254900				
H	8.76023300	1.30909400	0.24752000				
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H	11.09145700	2.86546600	-4.56050100				
H	11.88555700	1.63847600	-3.56440900				
H	11.81061600	3.30858700	-3.00537300				
C	-0.54976400	1.31239700	2.24967000				
C	0.07477900	2.31756100	1.48458800				
H	0.99401700	2.10793800	0.95271100				
C	-0.49790000	3.58122500	1.38320400	N	-2.62280200	0.08408500	0.04608000
H	0.00501800	4.33239400	0.78466800	C	-3.51063900	-0.99550500	0.31771900
C	-1.70918100	3.88817700	2.03946100	C	-3.34824900	-1.77101000	1.46943700
C	-2.32653600	2.87590500	2.80861300	H	-2.53591400	-1.55008200	2.15356500
H	-3.27733900	3.06175900	3.29074400	C	-4.22700300	-2.81695300	1.73246800
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C	-2.30101300	5.21982500	1.89157800	C	-5.45112700	-2.30243800	-0.27737500
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N	-1.16496000	8.86738800	-1.28148400	H	-3.02368600	3.82994100	-2.57555900
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4a

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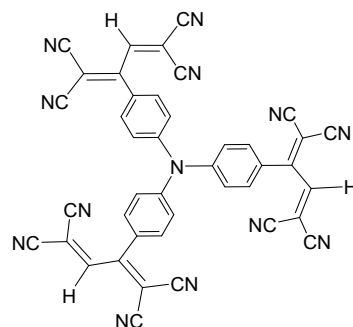
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C	5.27668000	2.07665600	1.68449300
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H	-5.96932200	-3.89791500	1.07334700
H	-4.79418800	4.77965900	-1.11467700



4b

N	0.01895300	1.81126800	0.12980600
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H	1.66803300	5.82999300	-1.42722200
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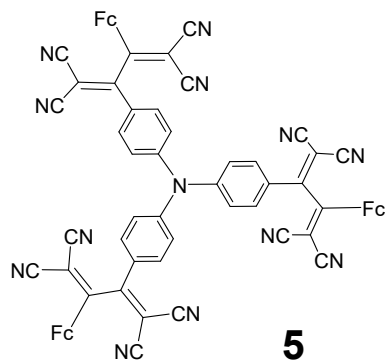
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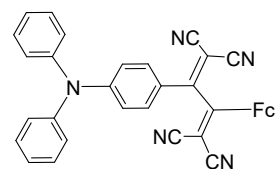


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N14	-2.2321	7.9824	2.8156
N15	1.8255	6.2940	3.3371
N16	1.9482	8.3600	-0.4887
C17	-2.8656	-1.9325	-0.9882
H18	-3.6369	-1.5884	-1.6706
C19	-1.8549	-1.0554	-0.6280
H20	-1.8489	-0.0465	-1.0284
C21	-0.8327	-1.4688	0.2423
C22	-0.8493	-2.7865	0.7294
H23	-0.0847	-3.1159	1.4259
C24	-1.8481	-3.6666	0.3434
H25	-1.8447	-4.6656	0.7654
C26	-2.8779	-3.2623	-0.5268
C27	-3.9627	-4.1582	-0.9362
C28	-3.8395	-5.5123	-1.0948
C29	-2.6015	-6.2267	-0.9873
C30	-4.9660	-6.3421	-1.4089
C31	-5.2958	-3.5239	-1.1779
C32	-5.7196	-3.4016	-2.4785
C33	-4.9077	-3.8254	-3.5799
C34	-6.9778	-2.8316	-2.8492
C35	-7.0141	-2.0118	0.0882
H36	-7.4974	-1.5114	-0.7379
C37	-5.9669	-3.0065	-0.0020
C38	-5.6518	-3.4117	1.3480
H39	-4.9339	-4.1715	1.6281
C40	-6.4614	-2.6629	2.2386
H41	-6.4787	-2.7653	3.3157
C42	-7.2913	-1.7957	1.4631
H43	-8.0439	-1.1217	1.8514

Supporting Information

C44	-8.0144	-5.7746	1.1285
H45	-7.2678	-6.5355	1.3149
C46	-8.7278	-5.0279	2.1147
H47	-8.6080	-5.1113	3.1875
C48	-9.5855	-4.1087	1.4370
H49	-10.2278	-3.3746	1.9068
C50	-9.4036	-4.2882	0.0309
H51	-9.8798	-3.7162	-0.7550
C52	-8.4371	-5.3192	-0.1565
H53	-8.0526	-5.6728	-1.1041
C54	1.5100	-1.0443	0.7913
C55	2.2970	-0.5979	1.8646
H56	1.8910	0.1161	2.5746
C57	3.5891	-1.0727	2.0340
H58	4.1692	-0.6975	2.8696
C59	4.1418	-2.0074	1.1391
C60	3.3530	-2.4256	0.0510
H61	3.7507	-3.1337	-0.6695
C62	2.0573	-1.9618	-0.1188
H63	1.4672	-2.3066	-0.9621
C64	5.5004	-2.5402	1.2892
C65	6.1385	-2.7338	2.4840
C66	5.5315	-2.5310	3.7665
C67	7.4987	-3.1838	2.5487
C68	6.2263	-2.8777	0.0258
C69	6.3595	-4.2029	-0.3050
C70	5.8019	-5.2407	0.5099
C71	7.0469	-4.6572	-1.4743
C72	6.6366	-1.7432	-0.7791
C73	6.8308	-0.4113	-0.2570
H74	6.7457	-0.1214	0.7825
C75	7.1872	0.4402	-1.3332
H76	7.4338	1.4908	-1.2533
C77	7.2226	-0.3431	-2.5280
H78	7.4921	0.0126	-3.5140
C79	6.9083	-1.6875	-2.1987
H80	6.8694	-2.5149	-2.8921
C81	10.0650	-1.1497	0.2524
H82	9.9485	-0.9295	1.3055
C83	10.3636	-0.2120	-0.7827
H84	10.5010	0.8545	-0.6565
C85	10.3923	-0.9190	-2.0234
H86	10.5535	-0.4809	-3.0003
C87	10.1110	-2.2942	-1.7562
H88	10.0146	-3.0832	-2.4908
C89	9.9139	-2.4342	-0.3511
H90	9.6527	-3.3455	0.1706
C91	-0.1099	0.7826	0.8639
C92	-1.3063	1.1552	1.4992
H93	-2.0108	0.3923	1.8155
C94	-1.5927	2.4902	1.7376
H95	-2.5308	2.7332	2.2228
C96	-0.6990	3.5071	1.3503
C97	0.4865	3.1222	0.6943
H98	1.1940	3.8767	0.3647
C99	0.7838	1.7887	0.4622
H100	1.7063	1.5213	-0.0435
C101	-0.9603	4.9303	1.5797
C102	-1.7173	5.4451	2.5987
C103	-2.3024	4.6677	3.6505
C104	-1.9832	6.8501	2.7042
C105	-0.3541	5.8841	0.5980

C106	0.7409	6.6086	0.9980
C107	1.3225	6.4371	2.2957
C108	1.3979	7.5689	0.1662
C109	-0.9394	5.8703	-0.7283
C110	-2.2686	5.3869	-1.0183
H111	-2.9873	5.0390	-0.2874
C112	-2.4790	5.4762	-2.4174
H113	-3.3933	5.2225	-2.9377
C114	-1.2971	6.0167	-3.0119
H115	-1.1570	6.2350	-4.0626
C116	-0.3540	6.2832	-1.9854
H117	0.6290	6.7081	-2.1272
C118	-2.0960	9.0328	-0.4441
H119	-1.5803	9.1363	0.5015
C120	-3.4150	8.5146	-0.6138
H121	-4.0661	8.1733	0.1804
C122	-3.7036	8.5000	-2.0122
H123	-4.6111	8.1298	-2.4722
C124	-2.5608	9.0057	-2.7036
H125	-2.4516	9.0840	-3.7779
C126	-1.5652	9.3340	-1.7325
H127	-0.5676	9.7011	-1.9365



6

1 Fe	4.1921	0.3038	-1.4411
2 N	-4.3421	-0.0040	-0.1839
3 N	0.4523	2.8807	3.8418
4 N	4.2255	1.1342	2.8779
5 N	1.5049	-2.3588	3.7168
6 N	4.0370	-3.5416	0.3903
7 C	-6.3508	-2.7626	-1.6676
8 H	-6.9427	-2.9493	-2.5596
9 C	-5.7240	-1.5274	-1.5061
10 H	-5.8272	-0.7540	-2.2619
11 C	-4.9585	-1.2757	-0.3609
12 C	-4.8292	-2.2689	0.6193
13 H	-4.2490	-2.0686	1.5158
14 C	-5.4466	-3.5064	0.4431
15 H	-5.3404	-4.2700	1.2089
16 C	-6.2115	-3.7592	-0.6985
17 C	-3.0094	0.0991	0.1901
18 C	-2.5259	1.2409	0.8674
19 H	-3.2037	2.0545	1.1022
20 C	-1.2049	1.3290	1.2621
21 H	-0.8968	2.2250	1.7862
22 C	-0.2791	0.2957	0.9928
23 C	-0.7677	-0.8262	0.2855
24 H	-0.0974	-1.6420	0.0337
25 C	-2.0905	-0.9337	-0.0970
26 H	-2.4220	-1.8158	-0.6339
27 C	1.1163	0.3308	1.3986
28 C	1.6835	1.1526	2.3473
29 C	0.9766	2.0953	3.1581

Supporting Information

30 C	3.0881	1.1206	2.6237	49 H	6.2564	-0.1144	-3.2840
31 C	2.0382	-0.6214	0.6955	50 C	5.9865	-0.6486	-1.1217
32 C	2.4115	-1.7634	1.3566	51 H	6.1051	-1.7244	-1.1132
33 C	1.9123	-2.0733	2.6631	52 C	5.6579	0.1621	0.0039
34 C	3.3074	-2.7358	0.8101	53 H	5.4898	-0.1823	1.0153
35 C	2.3751	-0.2766	-0.6737	54 C	-5.1435	1.1565	-0.3808
36 C	2.2891	1.0568	-1.2163	55 C	-6.4042	1.2466	0.2223
37 H	2.0323	1.9478	-0.6582	56 H	-6.7550	0.4354	0.8536
38 C	2.6227	1.0000	-2.5931	57 C	-7.1997	2.3719	0.0094
39 H	2.6826	1.8464	-3.2647	58 H	-8.1755	2.4340	0.4835
40 C	2.9224	-0.3580	-2.9232	59 C	-6.7441	3.4193	-0.7948
41 H	3.2419	-0.7219	-3.8912	60 C	-5.4856	3.3281	-1.3950
42 C	2.7965	-1.1469	-1.7497	61 H	-5.1254	4.1324	-2.0309
43 H	2.9720	-2.2108	-1.6823	62 C	-4.6902	2.2000	-1.1987
44 C	5.5256	1.5120	-0.4400	63 H	-3.7187	2.1226	-1.6792
45 H	5.2525	2.3544	0.1818	64 H	-7.3642	4.2970	-0.9541
46 C	5.7791	1.5380	-1.8452	65 H	-6.6972	-4.7221	-0.8297
47 H	5.7235	2.4081	-2.4873				
48 C	6.0633	0.2029	-2.2669				

8. Frontier MOs of 1'-6

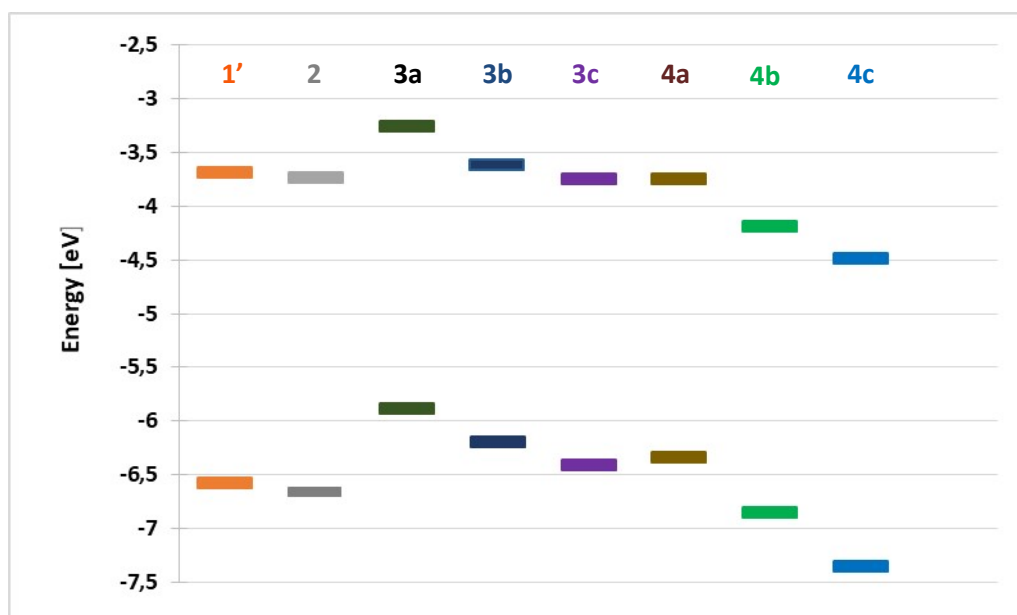
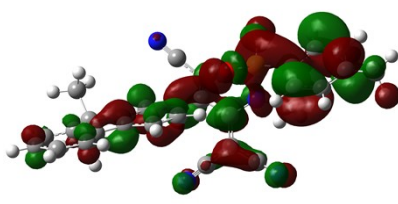
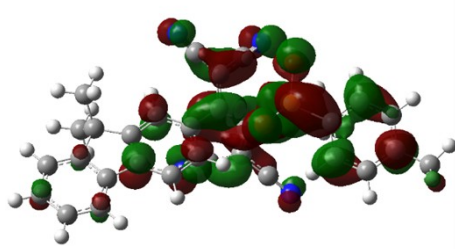
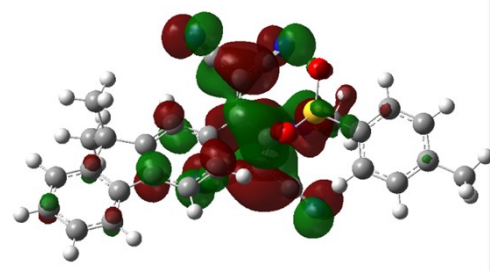
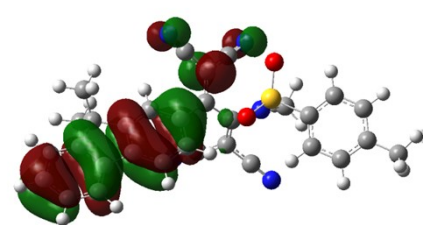
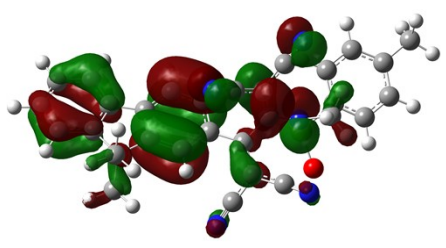
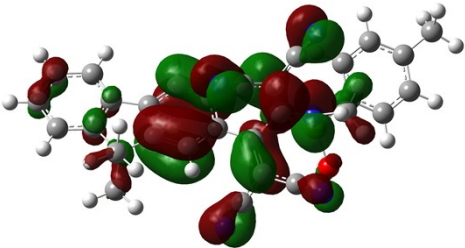
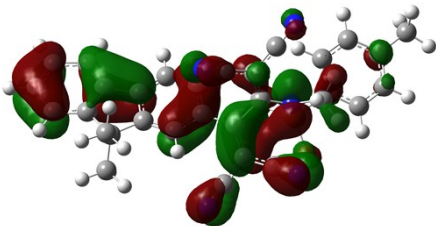
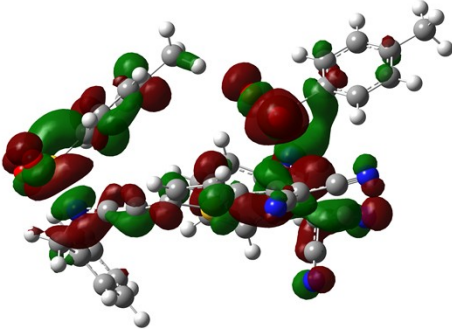
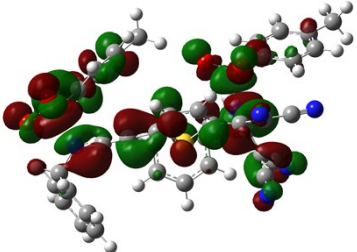
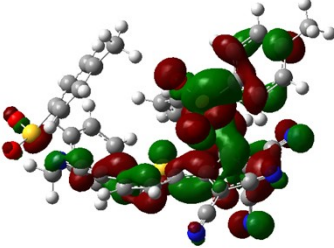


Figure S11. Energies of the HOMOs and LUMOs of the organic derivatives 1'-4c.

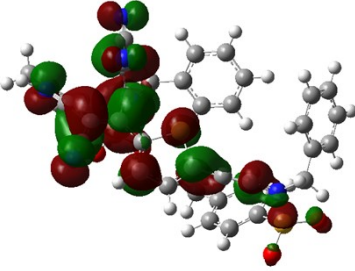
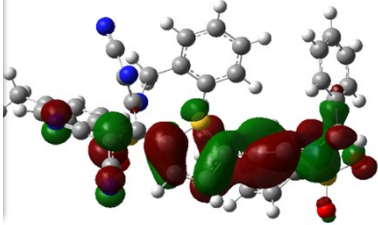
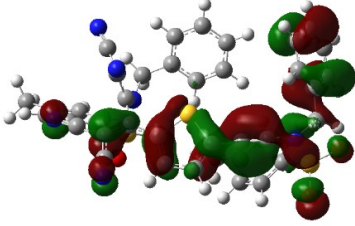
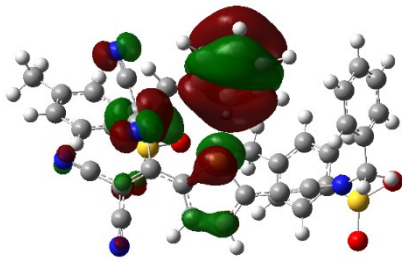
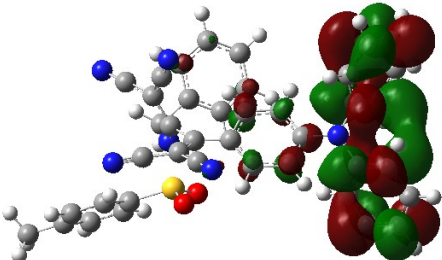
Supporting Information

1'	
<p>136 LUMO+1 (Ts/TCNE/Flu)</p>	
<p>135 LUMO +1 (TCNE/Ts)</p>	
<p>134 LUMO (TCNE)</p>	
<p>133 HOMO (FLUORENE)</p>	
<p>131 HOMO-2 (Flu/SO2)</p>	

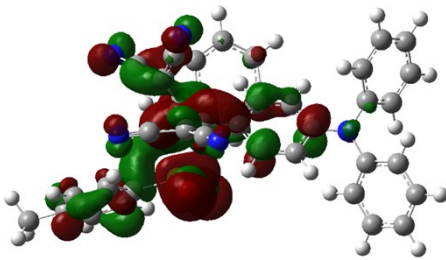
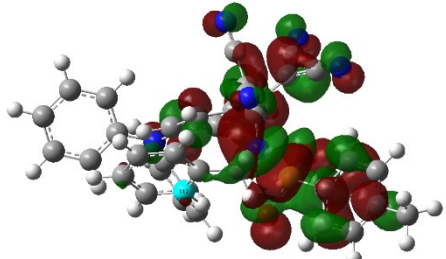
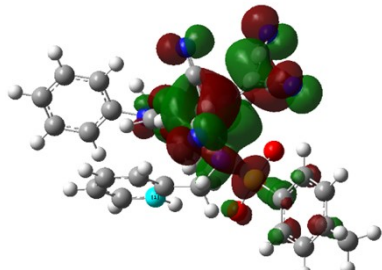
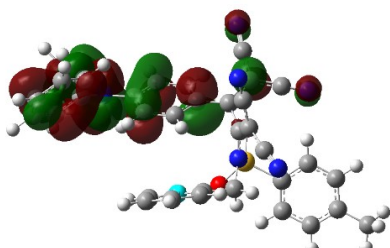
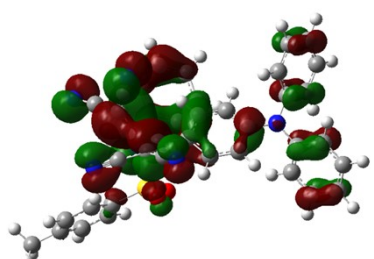
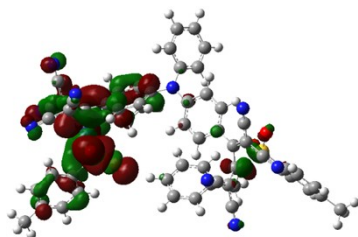
Supporting Information

<p>130 HOMO-3 (TCBD/SO₂)</p>	
<p>129 HOMO-4 (Flu/TCBD/SO₂)</p>	
2	
<p>191 LUMO +3 (2Ts/TCBD/CC)</p>	
<p>190 LUMO +2 (2Ts/TCBD/CC)</p>	
<p>189 LUMO +1 (TCNE/Ts/THIENYL)</p>	

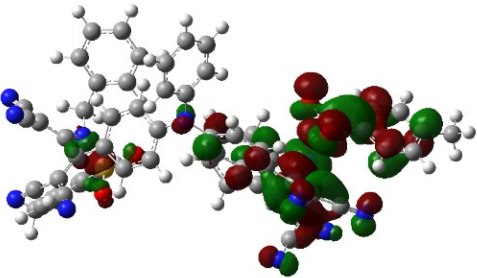
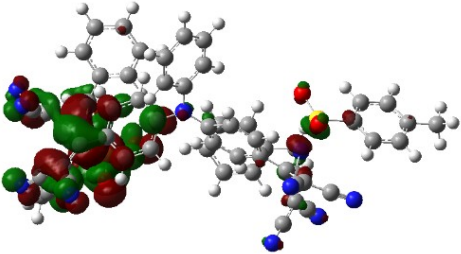
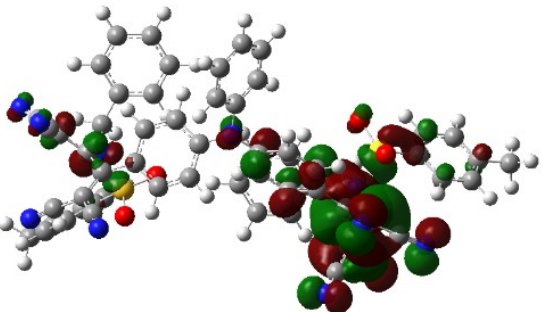
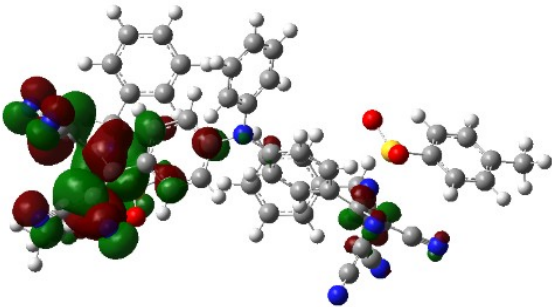
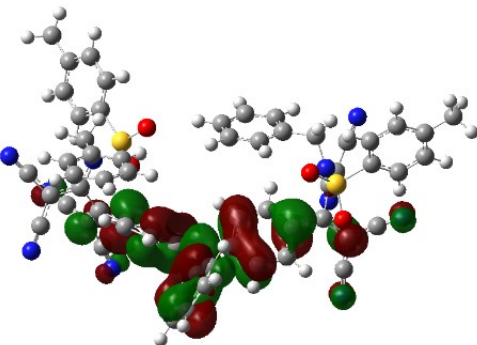
Supporting Information

<p>188 LUMO (TCNE)</p>	
<p>187 HOMO (THIENYL/C≡C)</p>	
<p>186 HOMO -1 (THIENYL/C≡C/PH)</p>	
<p>185 HOMO -2 (PH)</p>	
<p>3a</p>	
<p>173 LUMO+6 (NPH2)</p>	

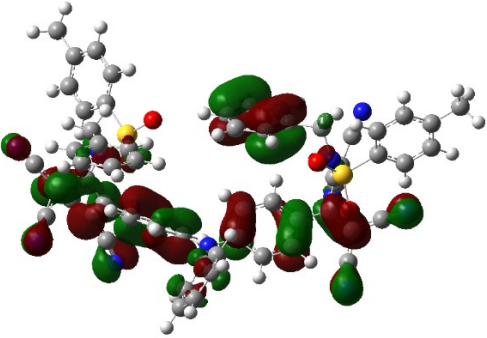
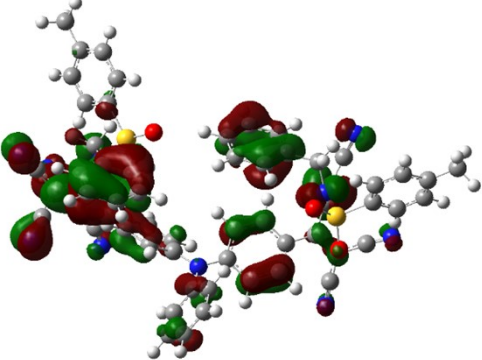
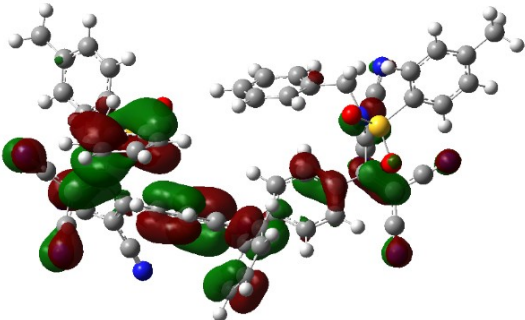
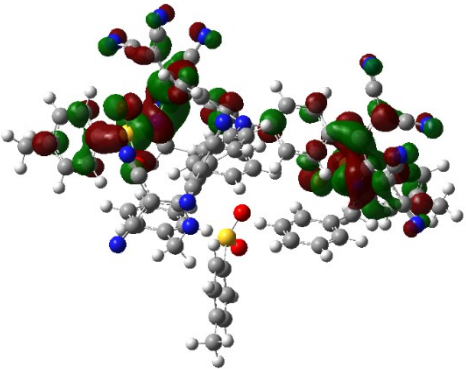
Supporting Information

<p>169 LUMO+2 (TCNE/Ts)</p>	
<p>168 LUMO+1 (TCNE/Ts)</p>	
<p>167 LUMO (TCNE)</p>	
<p>166 HOMO (NPH3)</p>	
<p>165 HOMO-1 (Bz/TCNE)</p>	
<p>3b</p>	
<p>272 LUMO+4 (TCNE/Ts)</p>	

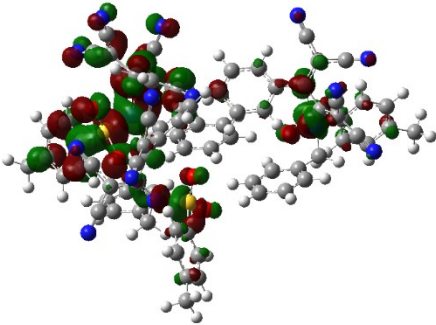
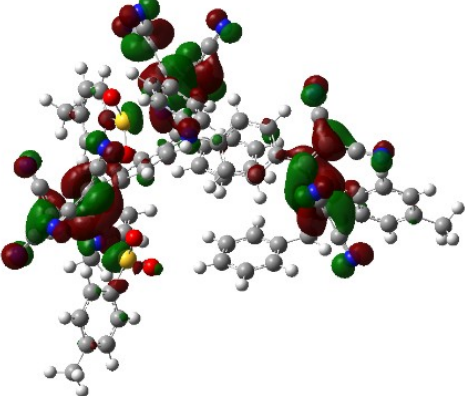
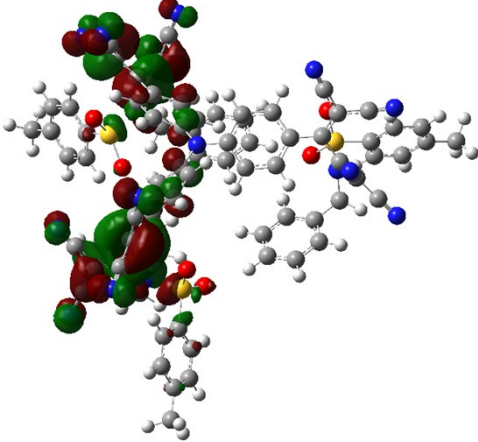
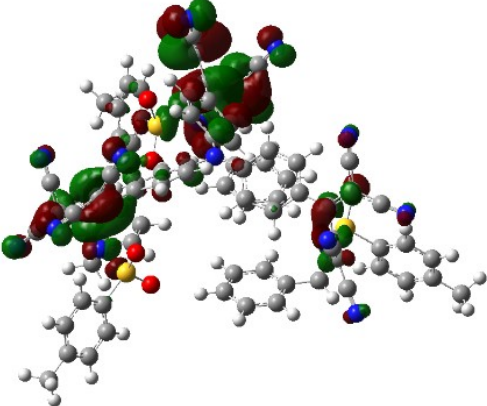
Supporting Information

<p>271 LUMO+3 (TCNE/Ts)</p>	
<p>270 LUMO+2 (TCNE/Ts)</p>	
<p>269 LUMO+1 (TCNE)</p>	
<p>268 LUMO (TCNE)</p>	
<p>267 HOMO (NPH2)</p>	

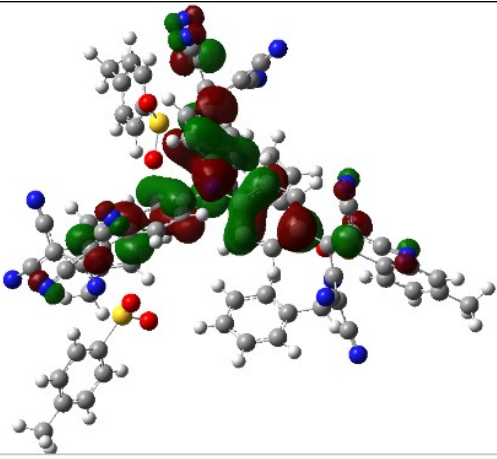
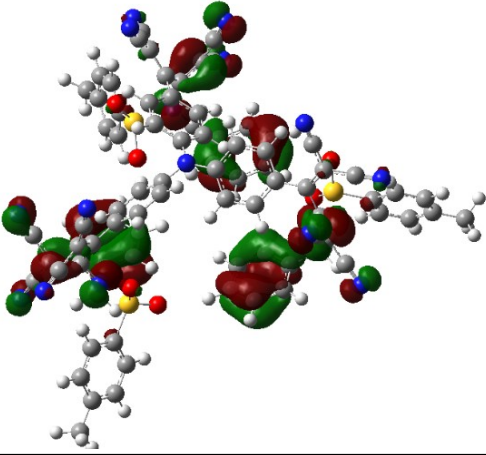
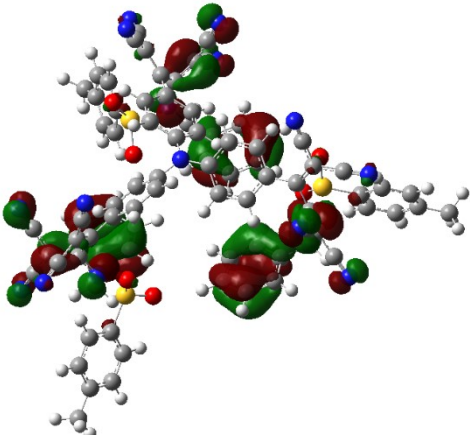
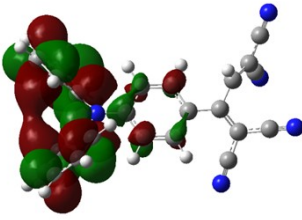
Supporting Information

<p>265 HOMO-2 (NPH₂/Bz/TCBD))</p>	
<p>264 HOMO-3 (NPH₂/Bz)</p>	
<p>261 HOMO-6 (Bz/NPH₂)</p>	
<p>3c</p>	
<p>373 LUMO+4 (TCNE/Ts)</p>	

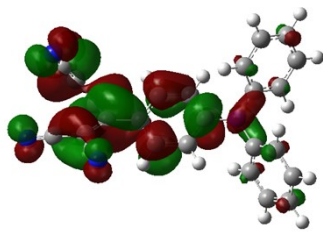
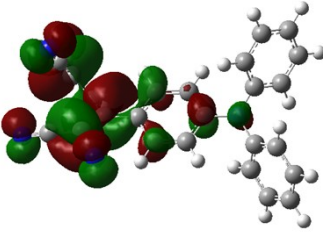
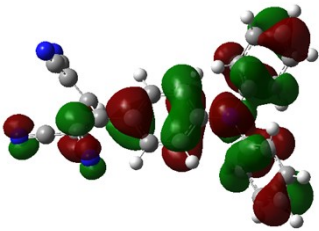
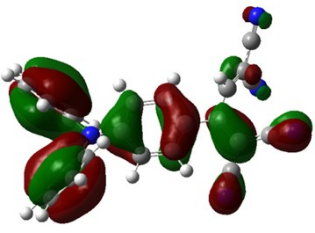
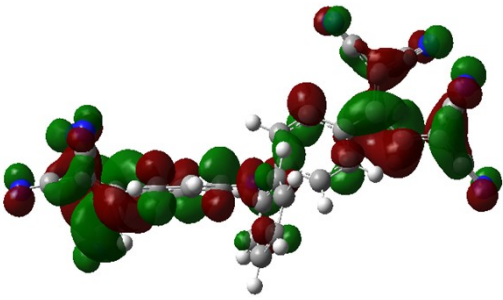
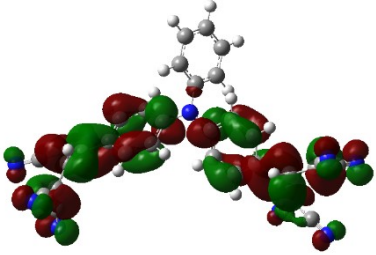
Supporting Information

<p>372 LUMO+3 (TCNE/Ts)</p>	
<p>371 LUMO+2 (TCNE)</p>	
<p>370 LUMO+1 (TCNE)</p>	
<p>369 LUMO (TCNE)</p>	

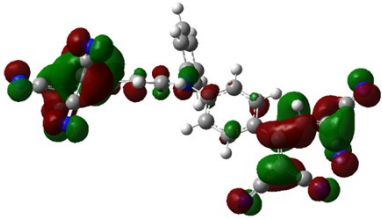
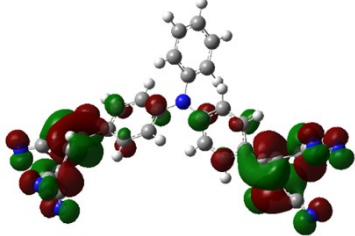
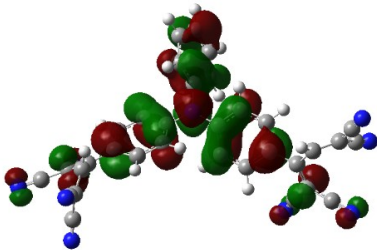
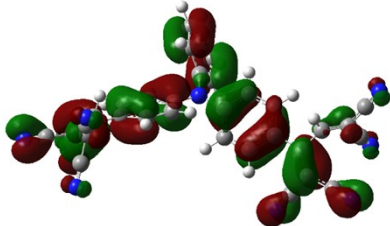
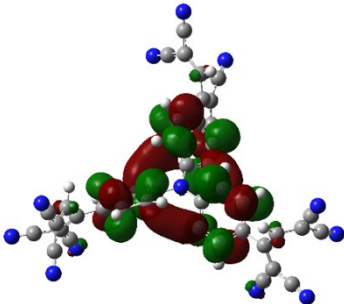
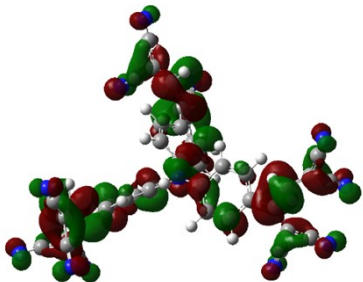
Supporting Information

<p>368 HOMO (NPH2)</p>	
<p>366 HOMO-2 (Bz)</p>	
<p>365 HOMO-3 (Bz)</p>	
<p>4a</p>	
<p>107 LUMO+3 (NPH2)</p>	

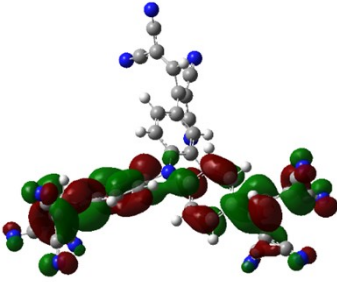
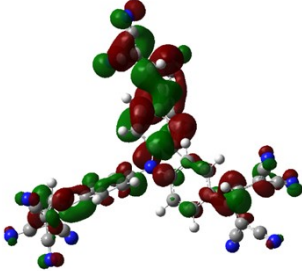
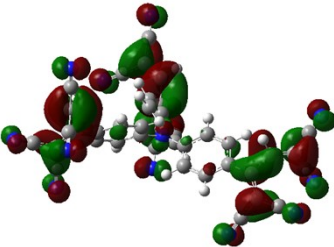
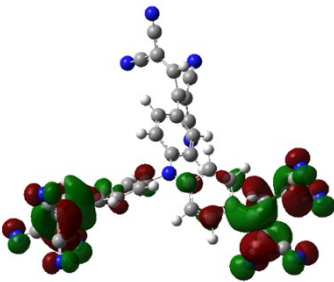
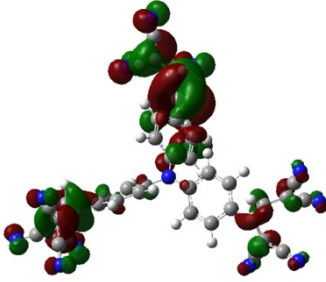
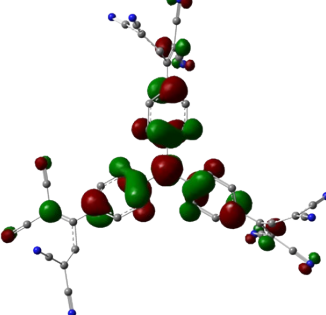
Supporting Information

<p>105 LUMO+1 (TCNE)</p>	
<p>104 LUMO (TCNE)</p>	
<p>103 HOMO (NPH3)</p>	
<p>102 HOMO-1 (NPH3/TCBD)</p>	
<p>4b</p>	
<p>145 LUMO+3 (TCNE/NPH2)</p>	
<p>144 LUMO+2 (TCNE/NPH2)</p>	

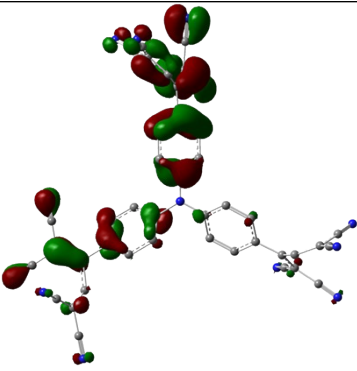
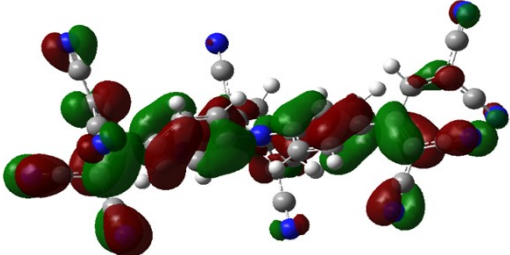
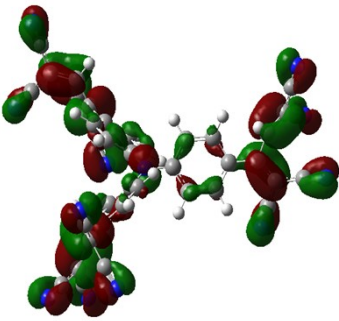
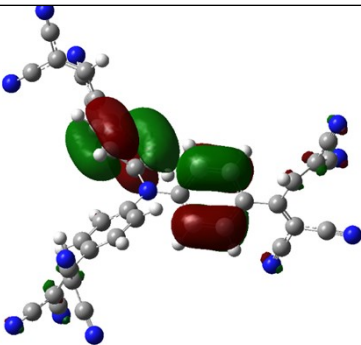
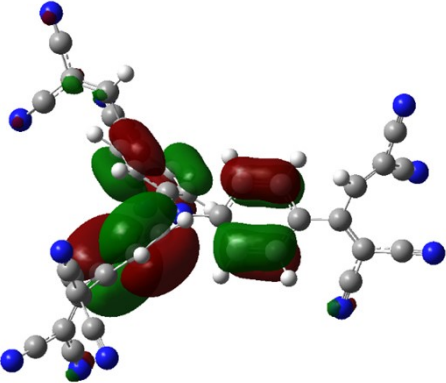
Supporting Information

<p>143 LUMO+1 (TCNE)</p>	
<p>142 LUMO (TCNE)</p>	
<p>141 HOMO (NPH3)</p>	
<p>140 HOMO-1 (NPH3/TCNE)</p>	
<p>4c</p>	
<p>186 LUMO+6 (TCNE)</p>	
<p>185 LUMO+5 (TCNE/ NPH3)</p>	

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<p>184 LUMO+4 (TCNE/PH)</p>	
<p>183 LUMO+3 (TCNE/PH)</p>	
<p>182 LUMO+2 (TCNE)</p>	
<p>181 LUMO+1 (TCNE)</p>	
<p>180 LUMO (TCNE)</p>	
<p>179 HOMO (NPH3)</p>	

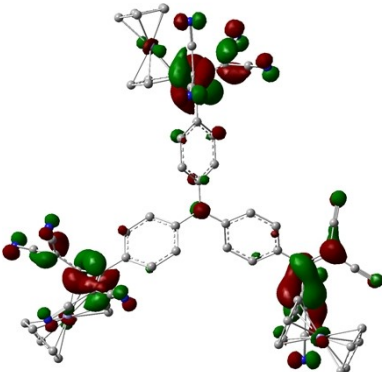
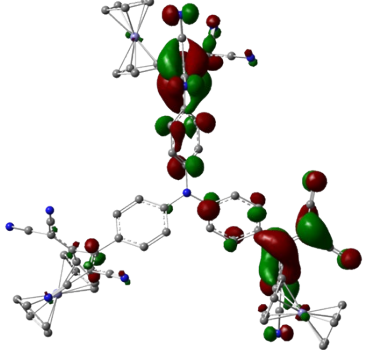
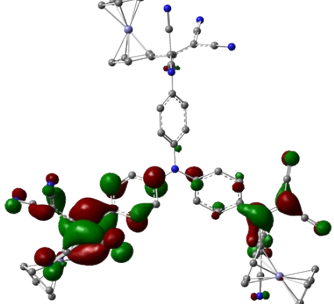
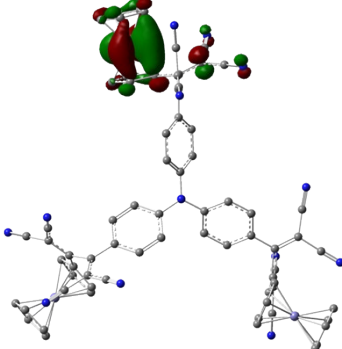
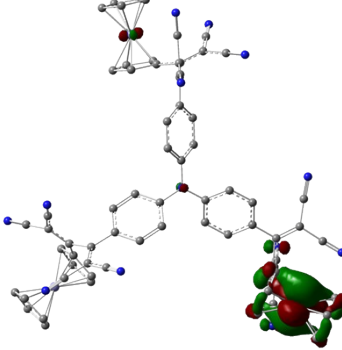
Supporting Information

<p>178 HOMO-1 (NPH3/TCBD)</p>	
<p>177 HOMO-2 (NPH3/TCBD)</p>	
<p>176 HOMO-3 (TCBD/NPH2)</p>	
<p>175 HOMO-4 (NPH2)</p>	
<p>174 HOMO-5 (NPH3)</p>	

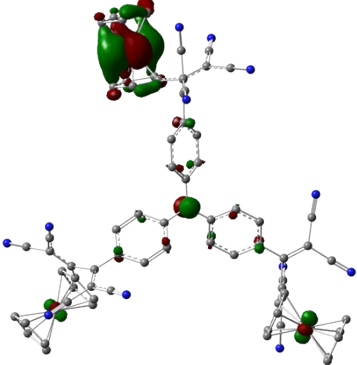
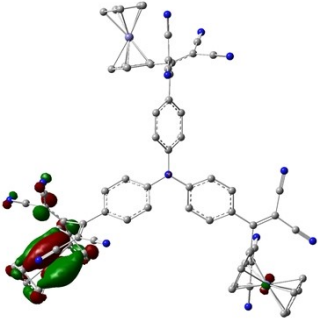
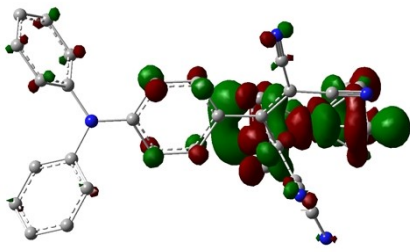
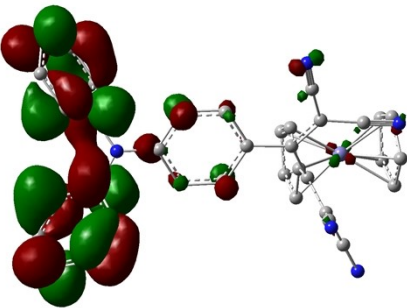
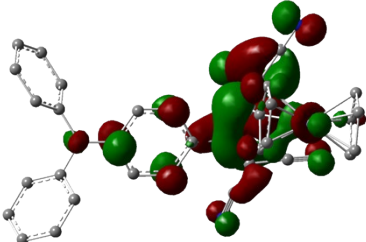
Supporting Information

5	
<p>328A LUMO+20 (Fc/TCBD)</p>	
<p>327A LUMO+20 (Fc/TCBD)</p>	
<p>326A LUMO+20 (Fc/TCBD)</p>	
<p>310A LUMO+4 (TCBD/Fc/PH)</p>	

Supporting Information

<p>308A LUMO+2 (TCBD/Fc)</p>	
<p>307A LUMO+1 (TCNE)</p>	
<p>306A LUMO (TCNE)</p>	
<p>305A HOMO (Fc)</p>	
<p>304A HOMO-1 (Fc)</p>	

Supporting Information

<p>303A HOMO-2 (Fc)</p>	
<p>302A HOMO-3 (Fc)</p>	
<p>6</p>	
<p>155A LUMO+9 (Fc)</p>	
<p>149A LUMO+3 (NPH3)</p>	
<p>147A LUMO+1 (TCBD)</p>	

Supporting Information

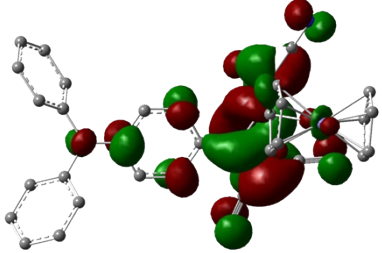
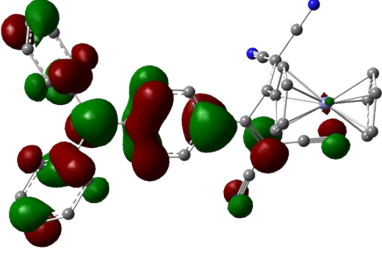
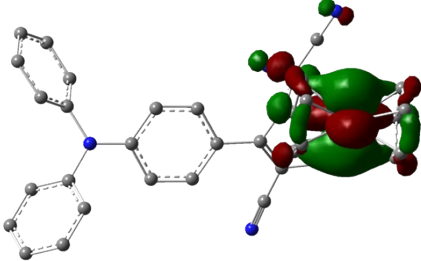
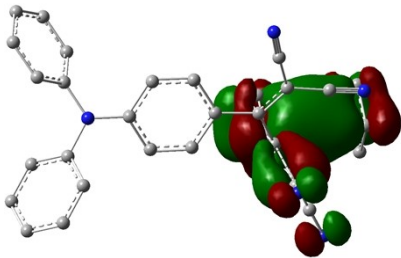
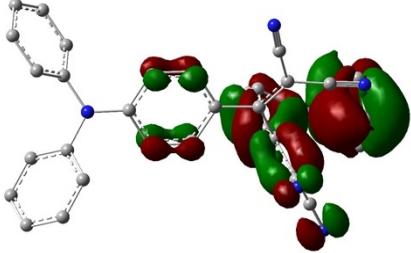
<p>146A LUMO (TCBD)</p>	
<p>145A HOMO (NPh3)</p>	
<p>144A HOMO-1 (Fc)</p>	
<p>142A HOMO-3 (Fc/TCBD)</p>	
<p>135A HOMO-10 (Fc/TCBD)</p>	

Figure S12. Frontier MOs of **1'-6**. Contour values are ± 0.02 (e/bohr³)^{1/2}.

9. Computed dipole moments of selected molecules

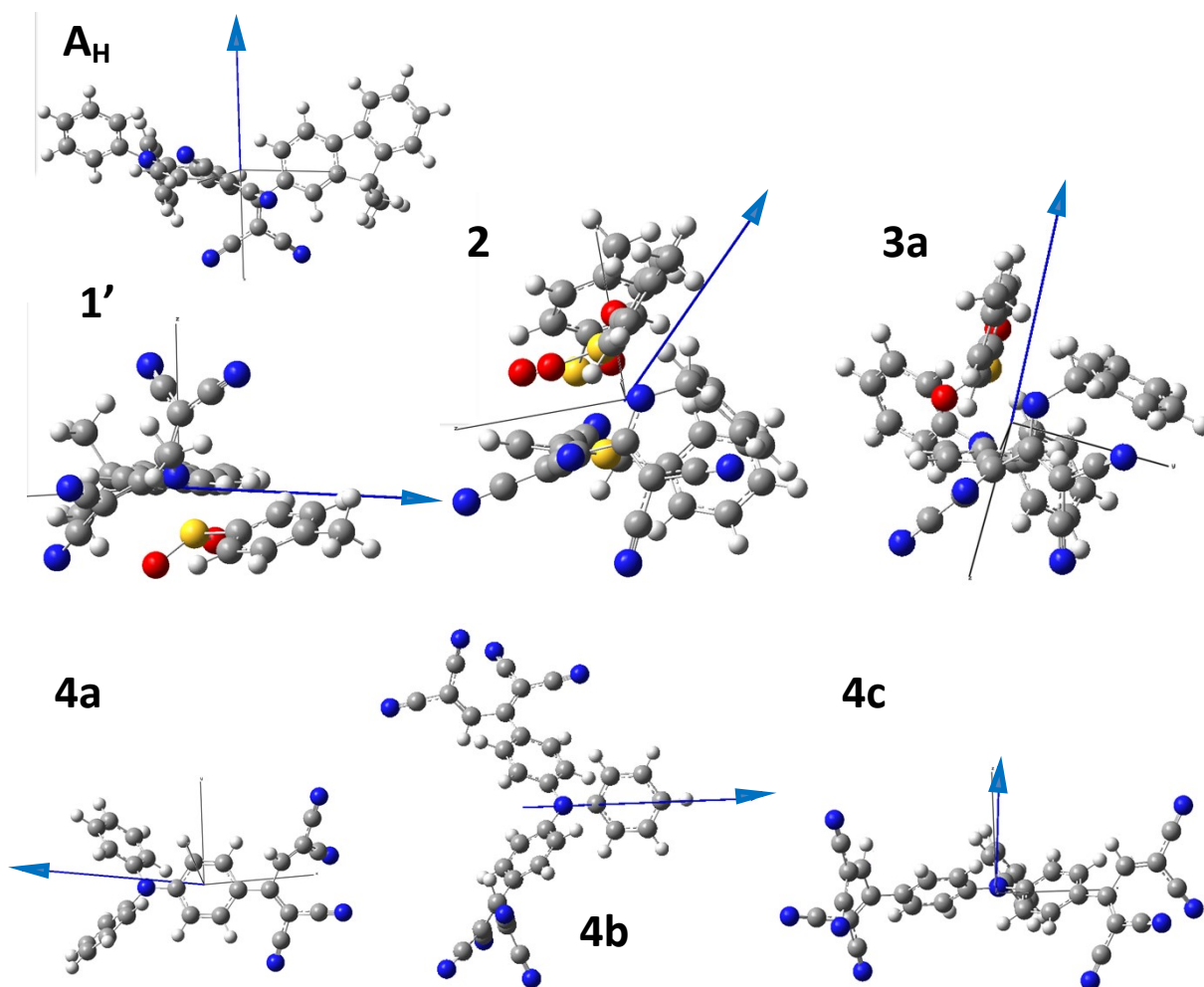


Figure S13. Graphical representation of the computed dipole moments for the organic derivatives A_H , $1'$ - 4 .

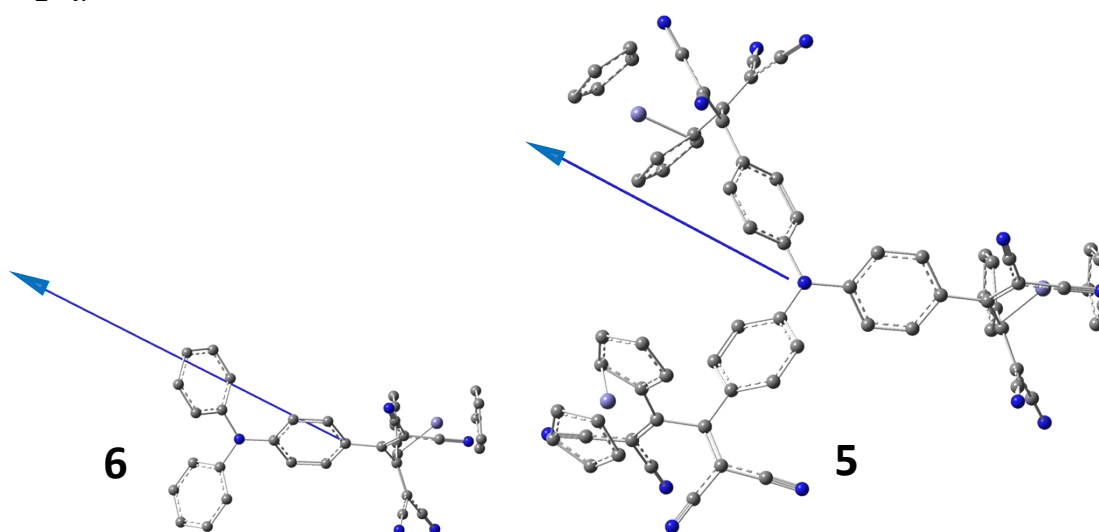


Figure S14. Graphical representation of the computed dipole moments for the organometallic derivatives 5 and 6 .

10. Computed isodensity-difference plots of selected molecules

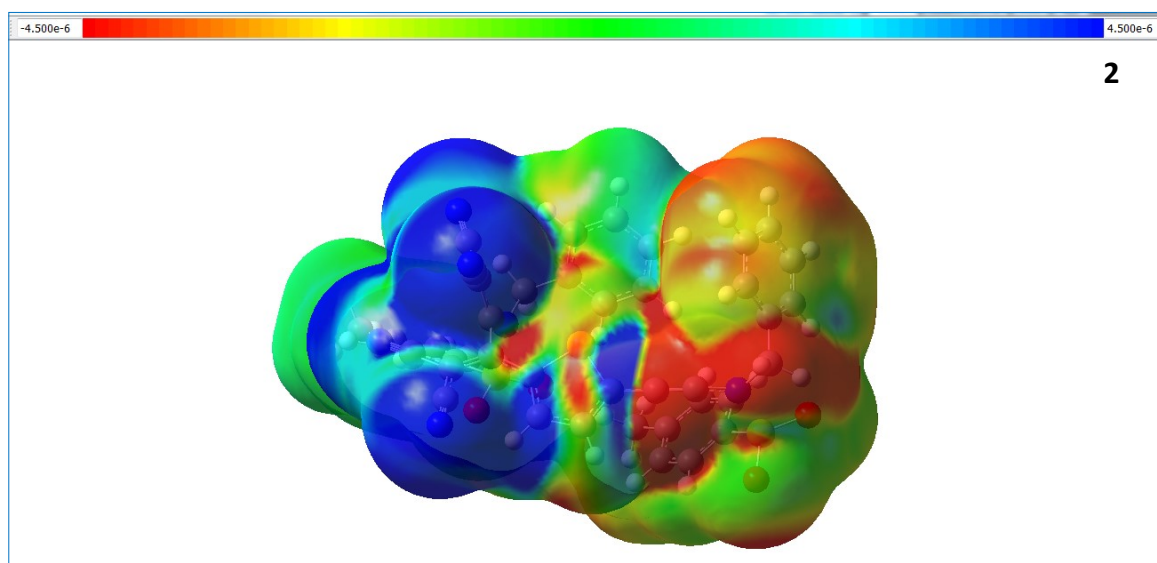
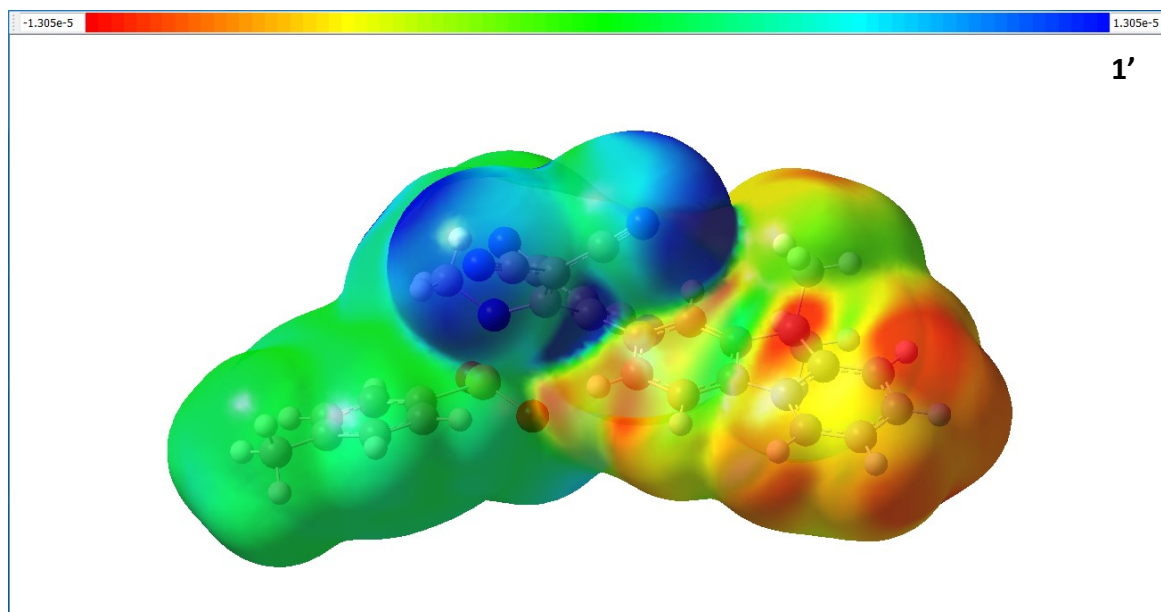


Figure S15. Density difference plots ($\Delta\rho(r) = \rho^{S_1}(r) - \rho^{S_0}(r)$) of **1'** and **2** between S_1 and S_0 states (red = decrease, blue = increase of electron density; isovalue 0.0001 au).

Supporting Information

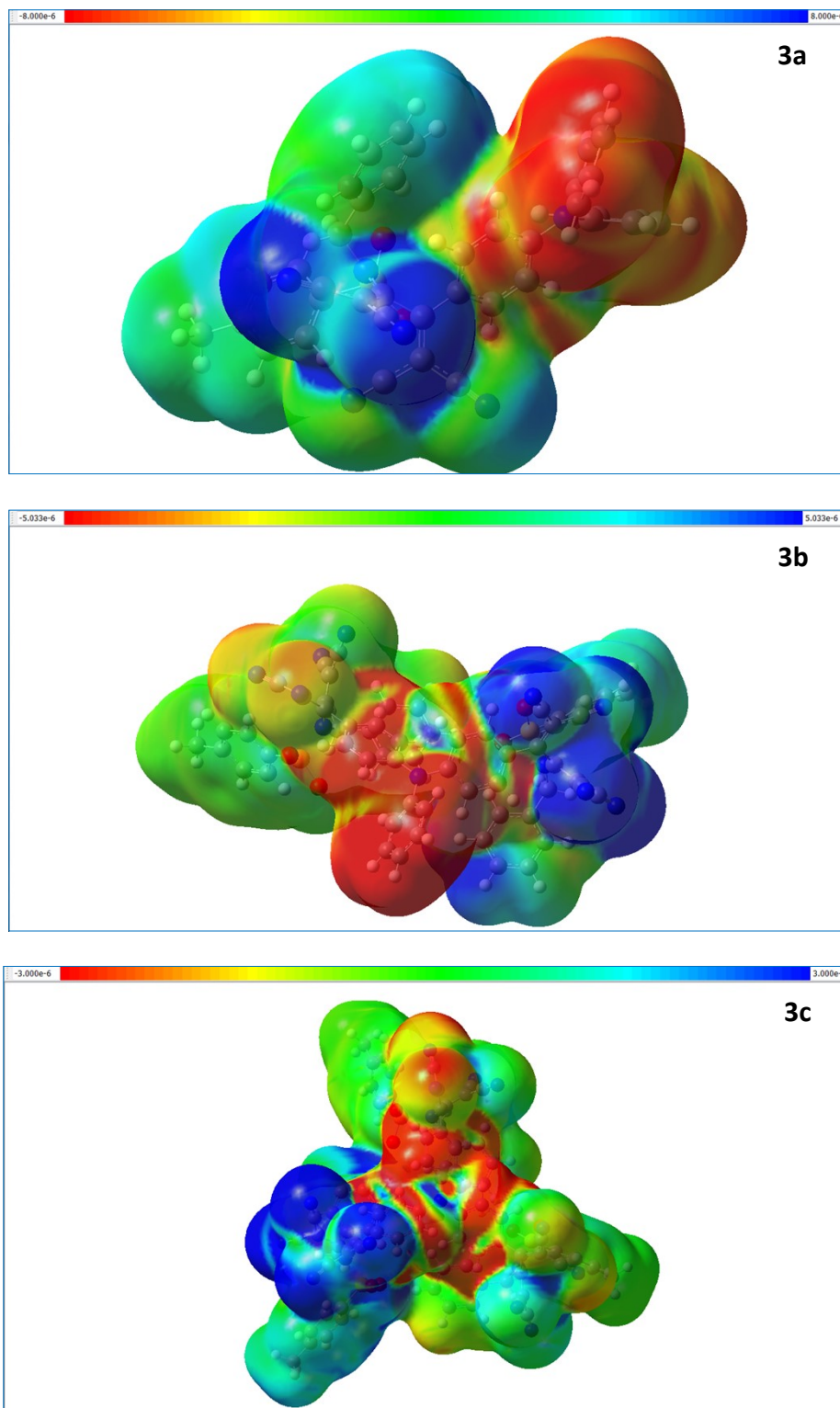


Figure S16. Density difference plots ($\Delta\rho(r) = \rho^{S_1}(r) - \rho^{S_0}(r)$) of **3a-c** between S_1 and S_0 states (red = decrease, blue = increase of electron density; isovalue 0.0001 au).

Supporting Information

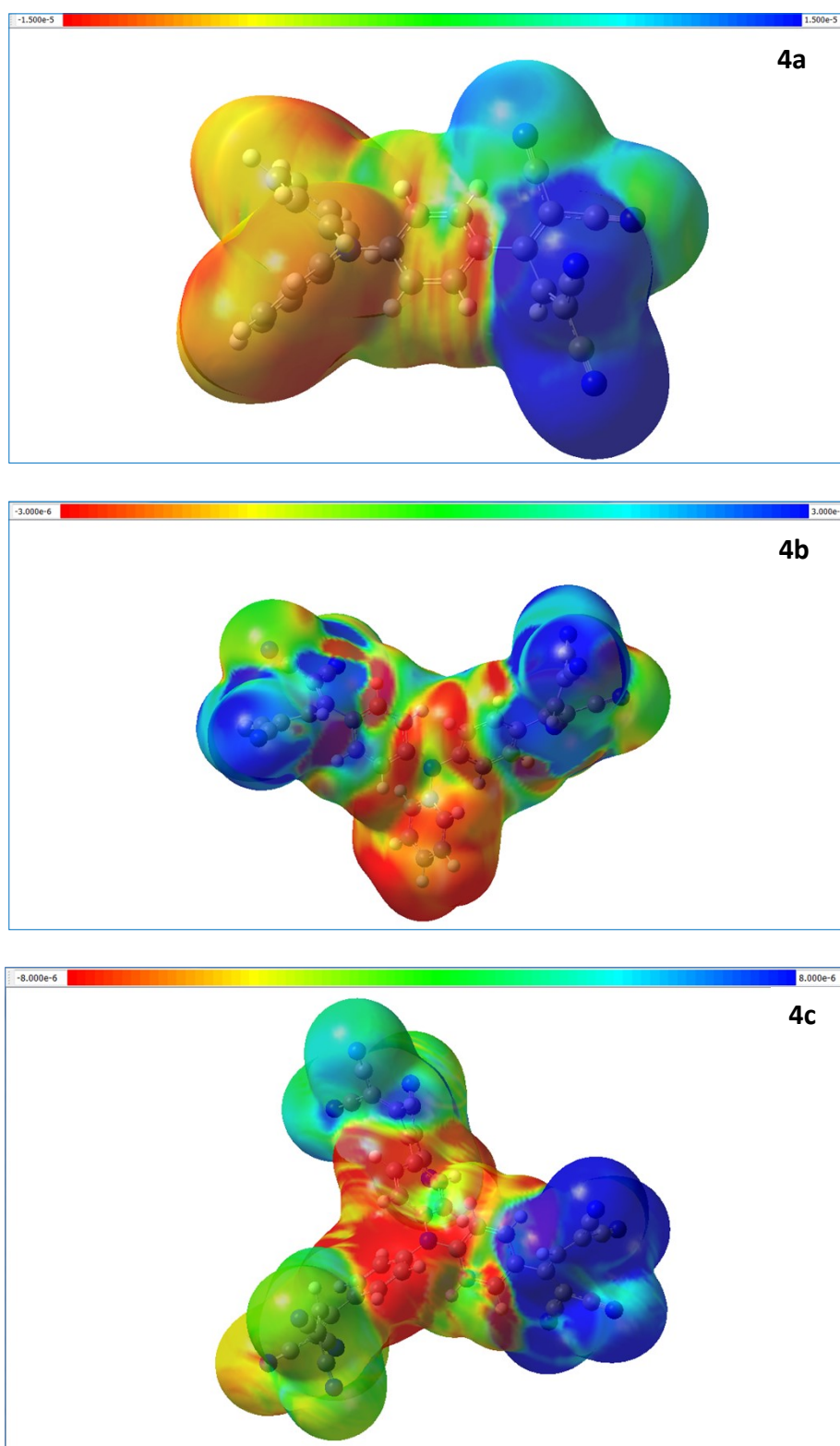


Figure S17. Density difference plots ($\Delta\rho(r) = \rho^{S_1}(r) - \rho^{S_0}(r)$) of **4a-c** between S_1 and S_0 states (red = decrease, blue = increase of electron density; isovalue 0.0001 au).

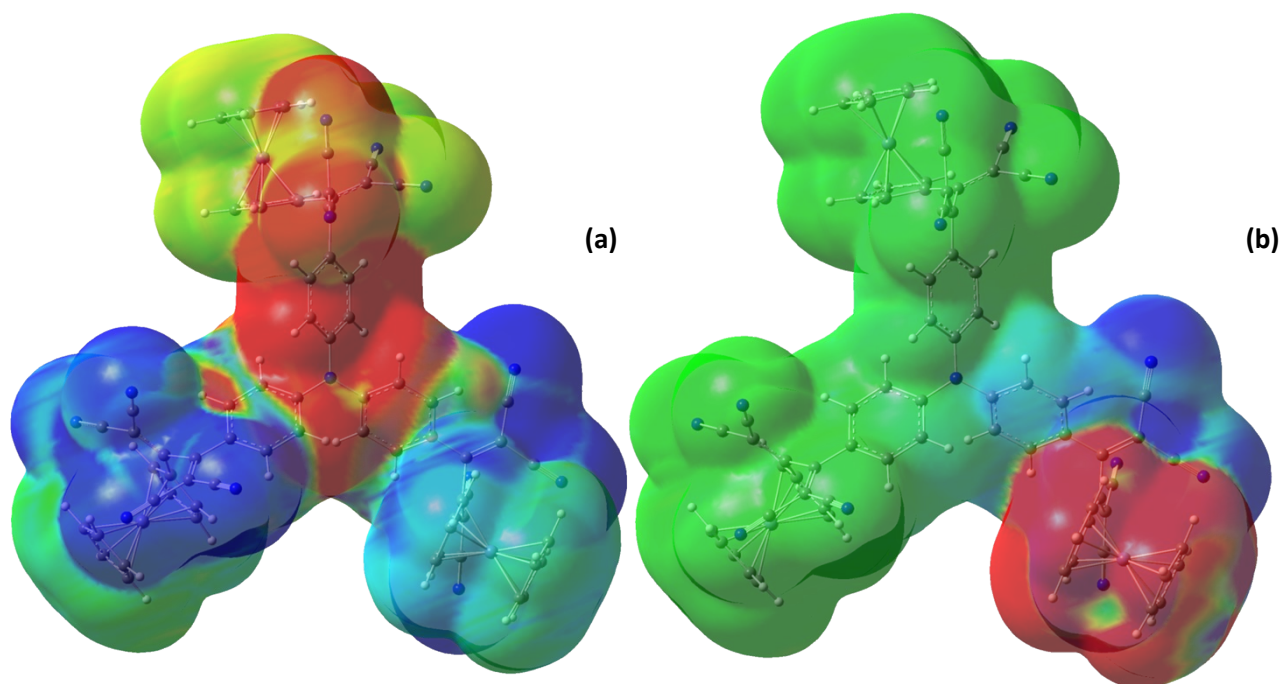


Figure S18. Density difference plots ($\Delta\rho(r)=\rho^{S_1}(r)-\rho^{S_0}(r)$) between S_1 and S_0 (a) or S_1 and S_2 states (b) (red = decrease, blue = increase of electron density; isovalue 0.0001 e/bohr³) for 5.

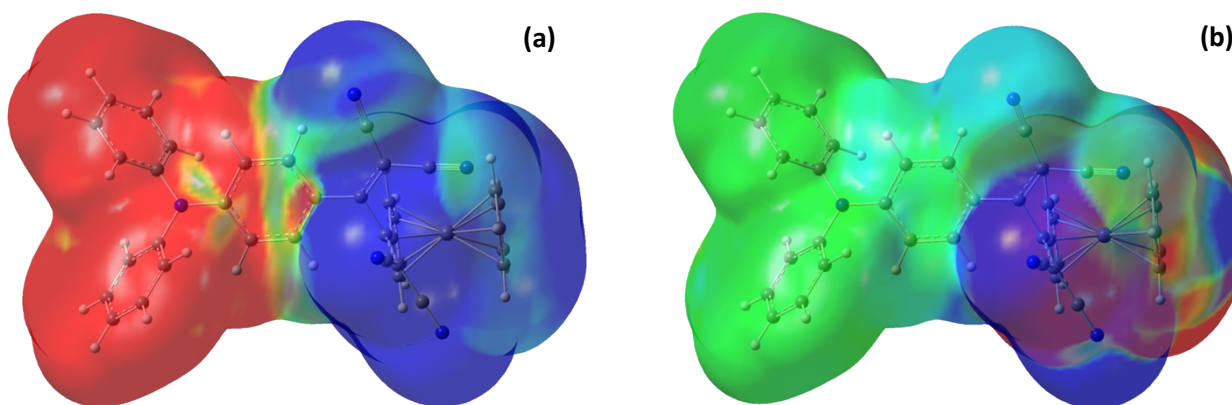


Figure S19. Density difference plots ($\Delta\rho(r)=\rho^{S_1}(r)-\rho^{S_0}(r)$) between S_1 and S_0 (a) or S_1 and S_2 states (b) (red = decrease, blue = increase of electron density; isovalue 0.0001 e/bohr³) for 6.

11. Solvent dielectric influence on computed transitions for 5

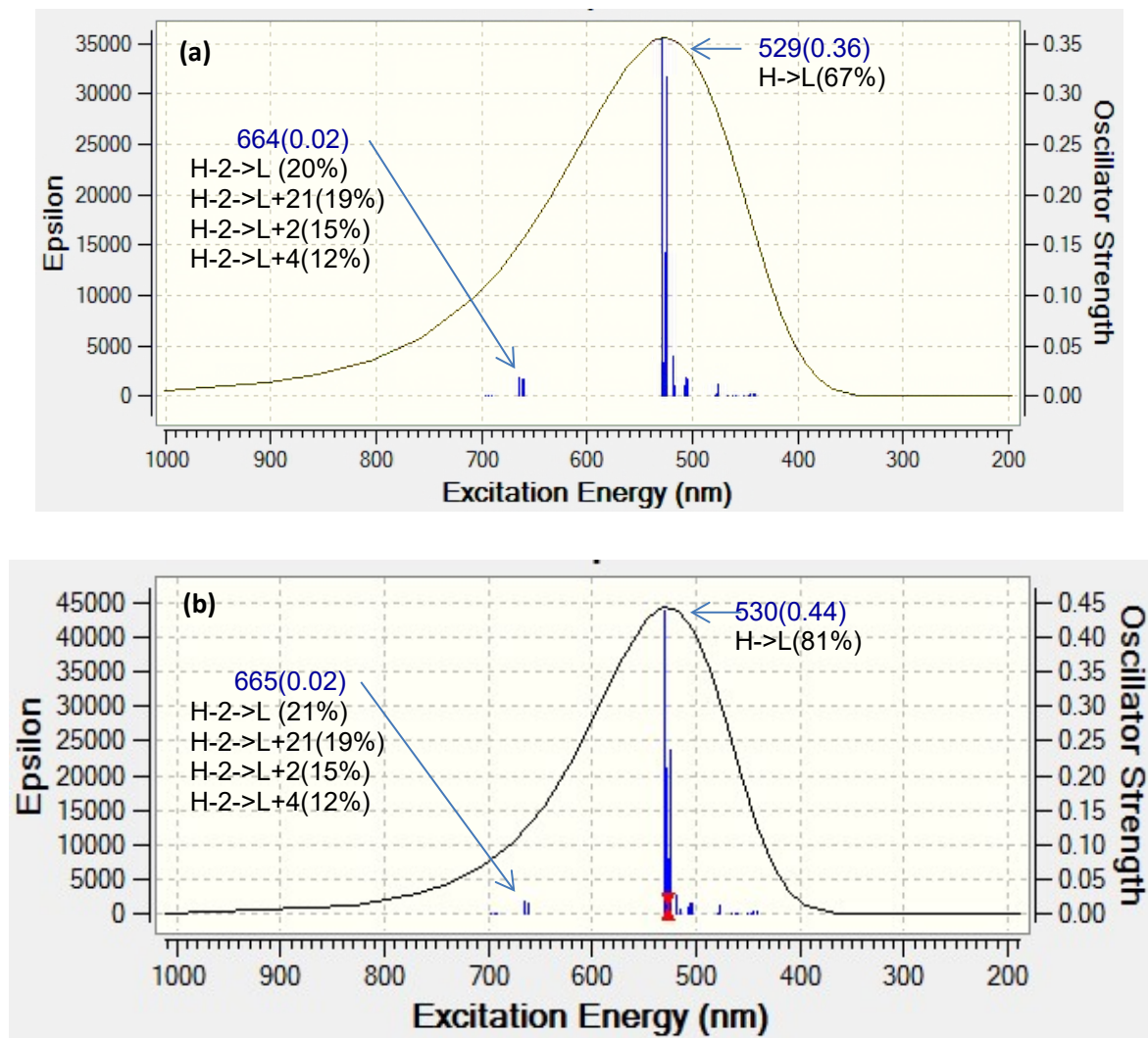


Figure S20. TD-DFT calculated spectrum for 5 (mPW1PW91/LANL2DZ) in THF (a) vs DCM (b).

12. Z-scan data for 1-5

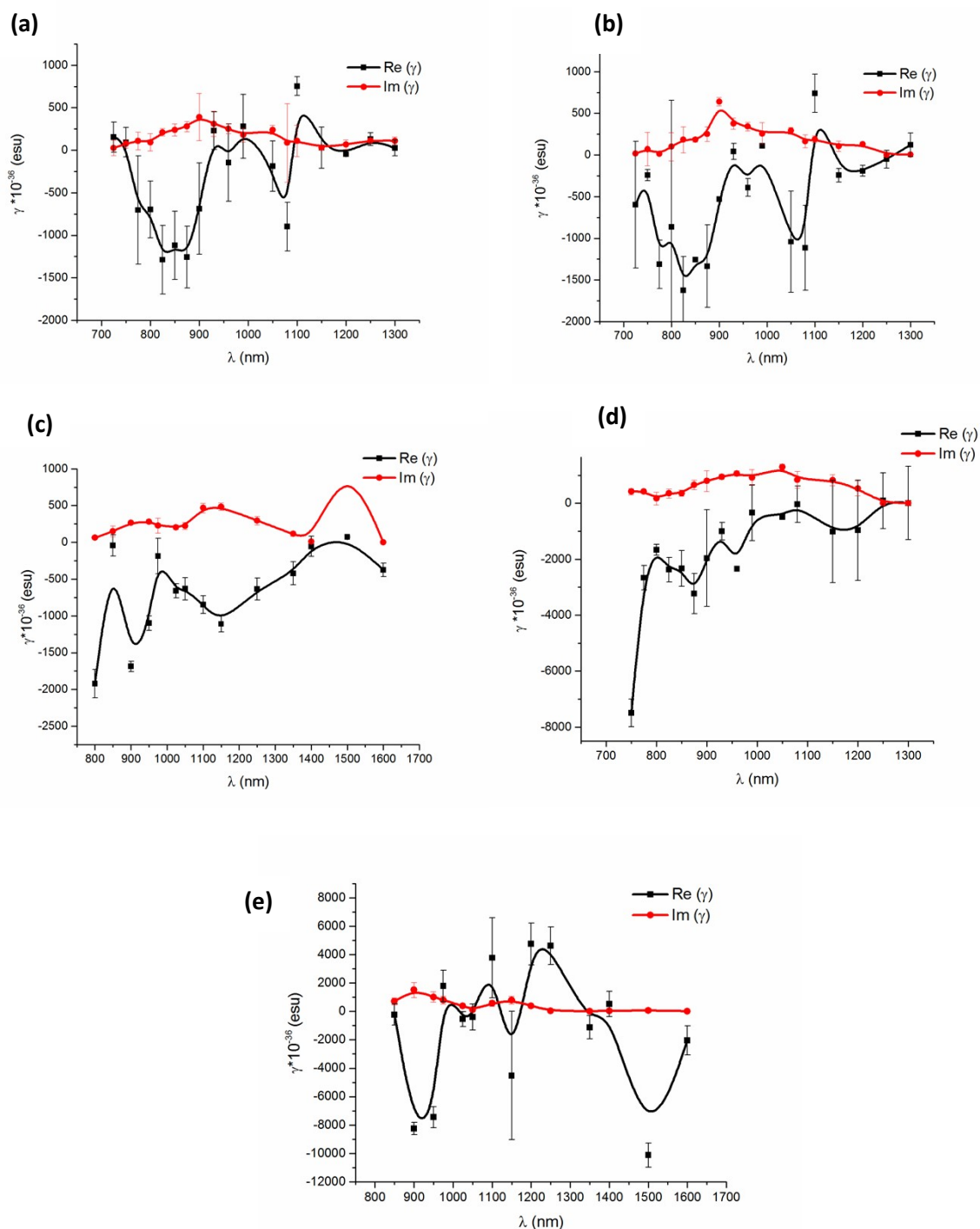


Figure S21. Overlays of real and imaginary cubic molecular nonlinear optical coefficients derived from closed- and open-aperture Z-scan measurements for **1** (a), **2** (b), **3a** (c), **3b** (d) and **3c** (e) in THF at 25 °C.

Supporting Information

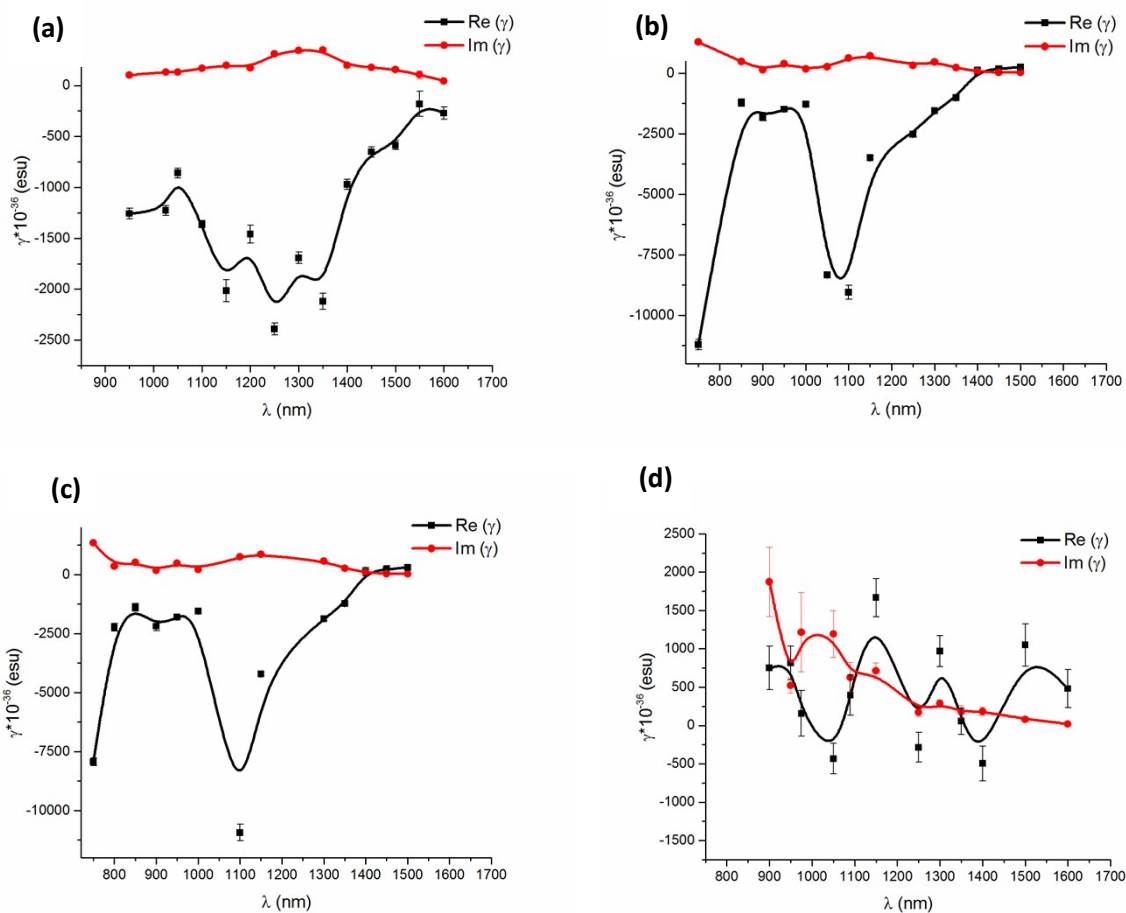


Figure S22. Overlays of real and imaginary cubic molecular nonlinear optical coefficients derived from closed- and open-aperture Z-scan measurements for **4a** (a), **4b** (b), **4c** (c) and **5** (d) recorded in CH_2Cl_2 (a) or THF (b-d) at 25 °C.

Table S2. Effective number of π electrons (N_{eff}) for compounds **1-5** and **A_x** (X = H, NPh₂).

Cmpd	N_{eff}
1	20.30
2	18.44
3a	18.44
3b	24.12
3c	28.35
4a	16.37
4b	20.68
4c	24.25
5	31.93
A_H	23.57
A_{NPh2}	25.05

Supporting Information

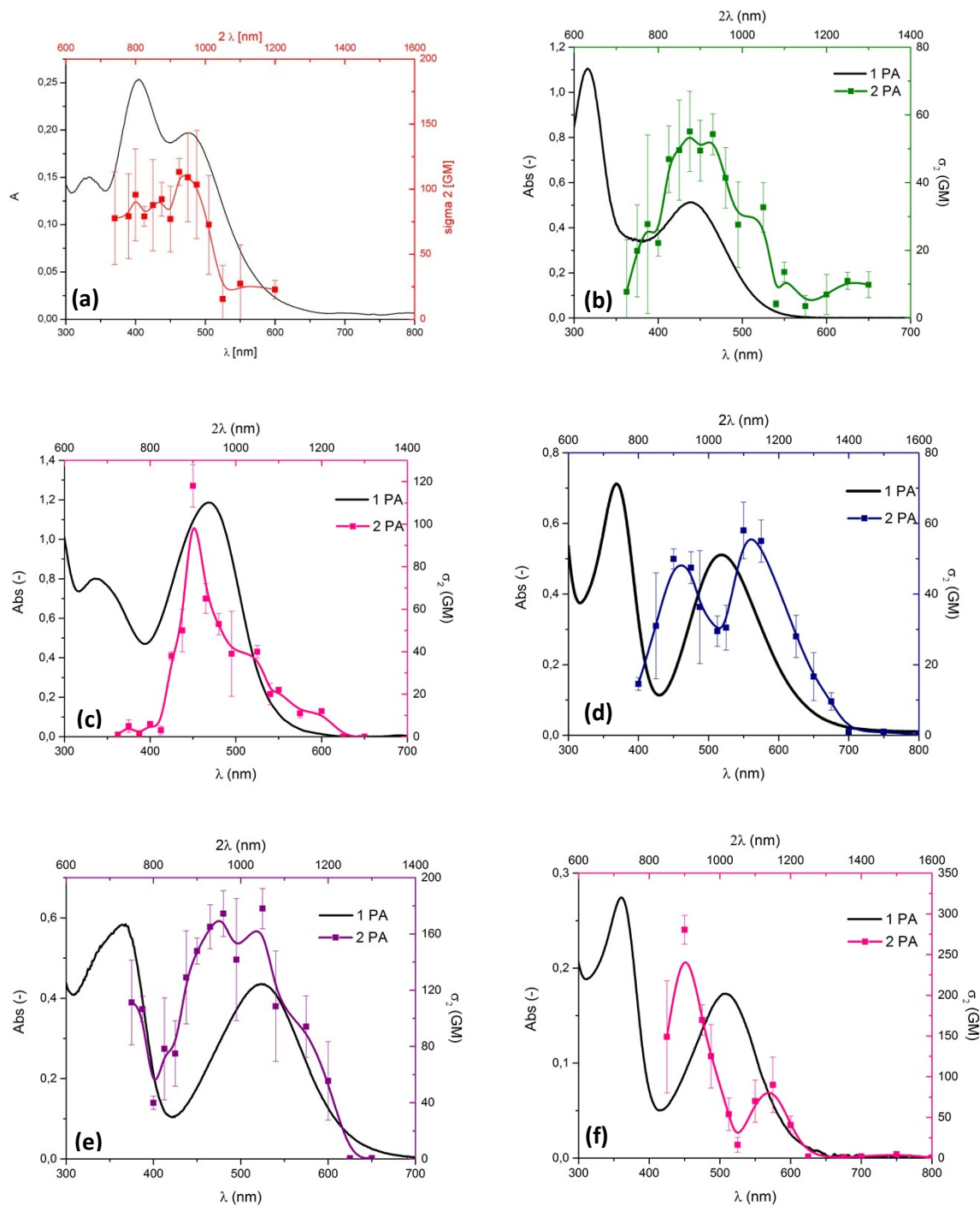


Figure S23. 2PA data for A_H and the tosylamide TCBD derivatives: Overlays of one- and two-photon absorption spectra for A_H (a), **1** (b), **2** (c), **3a** (d), **3b** (e), and **3c** (f). The two-photon cross-sections are derived from open-aperture Z-scan measurements recorded in THF at 25 °C. The one-photon spectra, recorded in CH_2Cl_2 (a-c) or THF (d-f), are plotted against twice the wavelength (2λ).

Supporting Information

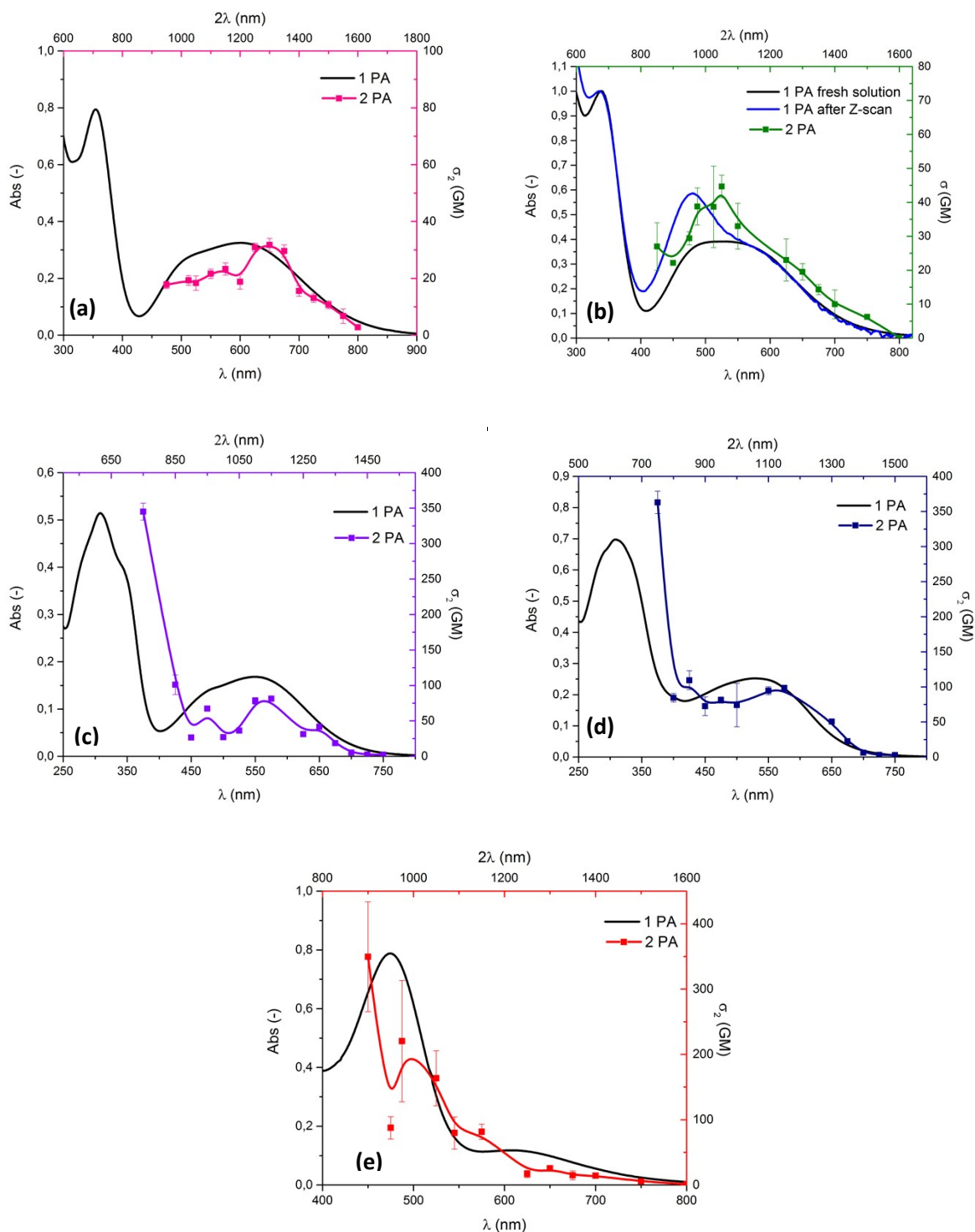


Figure S24. 2PA data for the other TCBD derivatives: Overlays of one- and two-photon absorption spectra for **4a** (a), **4a** (b), **4b** (c), **4c** (d), and **5** (e). The two-photon cross-sections are derived from open-aperture Z-scan measurements recorded in CH_2Cl_2 (a) or THF (b-e) at 25 °C. The one-photon spectra, recorded in CH_2Cl_2 (a) or THF (b-e), are plotted against twice the wavelength (2λ).