Supporting Information

Cascade Energy Transfer and Tunable Emission from Nanosheet Hybrids: Locating Acceptor Molecules Through Chiral Doping

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Materials

All chemicals, benzophenone, titanium tetrachloride, zinc powder, trimethylsilyl acetylene, copper iodide, bis(triphenylphosphine)palladium(II)dichloride, 1,3-dibromopropane, triethyl amine, dibromoanthracene, 4-iodophenol and potassium hydroxide were purchased from commercial suppliers and used as such without further purification. Dry solvents such as THF, DCM and acetone were purified prior to use according to the standard protocol and stored in molecular sieves.

General

All the reactions were carried out in oven dried round bottomed flasks under argon atmosphere unless otherwise mentioned. ¹H, ¹³C NMR spectra were recorded at Bruker-400/500 MHz NMR spectrometer instrument. The chemical shift values for ¹H and ¹³C NMR are recorded in CDCl₃ and TMS is used as internal standards for ¹H NMR. The value of coupling constant (J) is stated in Hertz (Hz). High-resolution mass spectra were recorded on JEOL JM AX 505 HA mass spectrometer. MALDI-TOF MS spectrum was recorded using DHB (2,5-dihydroxybenzoic acid) as the inert matrix on AB SCIEX MALDI TOF/TOF™ 5800. FT-IR spectra were recorded using Bruker Alpha FT-IR spectrometer and reported in frequency of absorption (cm⁻¹). UV-Vis spectra were recorded in shimadzu 1800 spectrophotometer, while all emission spectra were performed using PTI Quanta Master™ steady state spectrofluorometer. Fluorescence lifetimes were measured by time correlated single photon counting (TCSPC), using a spectroflourometer (Horiba scientific) and LED excitation source of 374 nm. The quality of the fit has been judged by the fitting parameters such as χ^2 (<1.2) as well as the visual inspection of the residuals. AFM imaging was performed on Key Sight 5500 instrument (Agilent Technologies) under tapping mode with silicon nitride tip. FEI Tecnai G2 F20 XTWIN TEM with accelerating voltage of 200 kV was used for the TEM imaging, 200 mesh TEM copper grid was purchased from TED PELLA, INC. SEM images were obtained using FEI, QUANTA 200 3D scanning electron microscope operating at 10, 15 and 20 kV using tungsten filament as electron source. Before imaging, the samples were sputtered with gold by using SCD 040 Balzers Union sputterer. Confocal laser scanning microscopy (CLSM) images were acquired in Olympus Fluoview Microscope. The Powder X-ray diffraction (PXRD) patterns were recorded on a Rigaku, MicroMax-007HF equipped with a high intensity Micro focus rotating anode X-ray generator. The data was collected with the help of Control Win software. A Rigaku, R-axis IV++ detector was used

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for the wide-angle experiments using Cu K (1.54 Å) radiation outfitted with a Ni filter and Aluminium holder was used as sample holder. Circular dichroism (CD) spectra were recorded on JASCO, J-815 spectropolarimeter. Thin film fluorescence quantum yield was measured using a Quanta-Phi 6" model F-3029.

Energy transfer and lifetime measurements

All the energy transfer and lifetime measurements were carried out in a concentration of 1 mM of **DADT** using a cuvette of path length 1 mm. Energy transfer studies in different concentrations of **DADT** (eg. 1×10^{-4} and 1×10^{-5} M) exhibited consistent results.

Synthesis

DADT is synthesized according to the following Schemes.

Scheme S1.



Scheme S2.



Synthesis Procedure

1,1,2-Triphenyl-2-(*p*-hydroxyphenyl)ethylene (3)^{S1}



Under argon atmosphere, a three necked flask was charged with zinc powder (14 g, 219 mmol) and 200 mL THF. The mixture was cooled to -5 to 0 °C, and TiCl₄ (12 mL, 109.76 mmol) was slowly added by keeping the temperature under 10 °C. The resulting suspension was warmed to room temperature and stirred for 0.5 hours, then heated to reflux for 2.5 hours. The mixture was again cooled to -5 to 0 °C, charged with pyridine (5 mL, 54.88 mmol) and stirred for 10 min. The solution of two carbonyl compounds (1:1 ratio, 4 g, 21.95 mmol) in 30 mL THF was added slowly. After addition, the reaction mixture was heated to reflux until the carbonyl compounds were consumed. The progress of the reaction was monitored by TLC (ethyl acetate/hexane, 2:8). The reaction was quenched with 10% K₂CO₃ aqueous solution and washed with DCM. The organic layer was separated, dried over anhydrous sodium sulphate. After removal of DCM *in vacuo*, the residue was purified by silica column chromatography using hexane and ethyl acetate as eluent, affording the products as a white solid. Yield: 52%.

FTIR (v_{max} in cm⁻¹): 3406-3681 (broad), 3019, 1595, 1215, 769. ¹H NMR (400 M Hz, CDCl₃), δ (TMS, ppm): 7.23 (d, J = 9.07, 2H), 7.30 (m, 15H), 6.84 (d, J = 8.76, 2H), 5.35 (s, 1H). ¹³C NMR (100 M Hz, CDCl₃), δ (ppm): 154.0, 143.9, 140.4, 136.3, 132.7, 131.3, 128.5, 126.2, 114.5. MALDI-TOF-MS: [M⁺] calcd. for C₂₆H₂₀O: 348.45, found: 349.32.

Synthesis of 4-(3-bromopropyloxy)iodobenzene (5)^{S2}



A mixture of 4-iodophenol (2 g, 9.09 mmol), 1,3-dibromopropane (12 mL, 181.81 mmol), potassium carbonate (5 g) in dry acetone (60 mL) in a two necked flask was refluxed at 60 °C under argon for 24 hours. The progress of the reaction was monitored by TLC (hexane/ethyl acetate, 19:1). After completion of the reaction, DCM and water was added to the reaction mixture. The organic layer was collected and dried over anhydrous sodium sulphate. After removal of DCM *in vacuo*, the residue was purified by silica column chromatography using hexane and ethyl acetate as eluent, affording the products as a white solid. Yield: 71%.

FTIR (v_{max} in cm⁻¹): 2925, 2854, 1660, 1239, 759. ¹H NMR (400 M Hz, CDCl₃), δ (TMS, ppm): 7.57 (d, J = 9.12, 2H), 6.7 (d, J = 8.82, 2H), 4.08 (t, J = 6.10, 2H), 3.60 (t, J = 6.22,2H), 2.32 (m, J = 6.03, 2H). ¹³C NMR (100 M Hz, CDCl₃), δ (ppm): 158.53, 138.23, 116.89, 82.97, 65.35, 32.15, 29.82. MALDI-TOF-MS: [M⁺] calcd. for C₉H₁₉BrI: 340.99, found: 341.97.

2-(4-(3-(4-Iodophenoxy)propoxy)phenyl)ethane-1,1,2-triyl)tribenzene (6)



A mixture of 1,1,2-triphenyl-2-(p-hydroxyphenyl)ethylene (500 mg, 1.43 mmol), 4-(3bromopropyloxy)-iodobenzene **5** (734 mg, 2.15 mmol), potassium carbonate (9.92 mg, 71.75 mmol), and dry acetone (25 mL) in a two necked flask was kept for reflux at 60 °C under argon for 24 hours. The progress of the reaction was monitored by TLC (ethyl acetate/hexane, 2:18). After completion of the reaction, DCM and water was added to the reaction mixture. The organic layer was separated, dried over anhydrous sodium sulphate. After removal of DCM *in vacuo*, the residue was purified by silica column chromatography using hexane and ethyl acetate as eluent, affording the products as a white solid. Yield: 82%. FTIR (v_{max} in cm⁻¹): 2924, 1660, 1216, 760. ¹H NMR (400 M Hz, CDCl₃), δ (TMS, ppm): 7.56 (d, J = 9.07, 2H), 7.07 (m, 15H), 6.94 (d, J = 8.76, 2H), 6.65(t, J = 8.8, 4H), 4.09 (m, 4H), 2.20 (t, J = 5.95, 2H). ¹³C NMR (100 M Hz, CDCl₃): δ (ppm): 158.7, 157.3, 143.9, 140.4, 138.2, 136.2, 132.5, 131.34, 127.7, 126.2, 116.9, 113.5, 82.7, 64.3, 31.9, 29.2.

9,10-Bis(ethynyl)anthracene (8)^{S3}

MALDI-TOF-MS: [M⁺] calcd. for C₃₅H₂₉IO₂: 608.52, found: 608.46.



A mixture of dibromoanthracene (3.2 g, 9.52 mmol), trimethylsilylacetylene (5 mL, 23.81 mmol), $PdCl_2(PPh_3)_2$ (669 mg, 9.52 mmol) was transferred to a two necked flask and dry THF (60 mL) was added. In another small two necked flask CuI was suspended in dry THF (3-4 mL). Both the flasks were degassed separately by repeated freeze-pump-thaw cycles. After this, CuI was mixed with the reaction mixture and kept for reflux at 70 °C under argon

for 24 hours. The progress of the reaction was monitored by TLC (hexane). The crude mixture was filtered through celite and washed with DCM. After removal of DCM *in vacuo*, the residue was purified by silica column chromatography using hexane as eluent, affording the products as a red solid. Yield: 50%.

FTIR (v_{max} in cm⁻¹): 3021, 2401, 1215, 758. ¹H NMR (400 M Hz, CDCl₃), δ (TMS, ppm): 8.58 (m, J = 3.42, 4H), 7.62 (m, = 3.24, 4H), 0.43 (s, 18H). ¹³C NMR (100 M Hz, CDCl₃), δ (ppm): 132.2, 127.1, 126.8, 118.4, 108.1, 101.4. 0.18. MALDI-TOF-MS: [M⁺] calcd. for C₂₄H₂₆Si: 370.64, found: 371.66.

9,10-diethynyl anthracene (9)^{S3}



9,10-bis(trimethylsilyl)ethynylanthracene (200 mg, 5.3 mmol) was dissolved in dry THF (20 mL) and the solution was degassed with argon for 15 minutes. Then KOH (151.37 mg, 2.70 mmol) in dry methanol was added. The reaction mixture was stirred at room temperature for 4 hours under argon atmosphere. The progress of the reaction was followed by TLC analysis and once the starting material was completely consumed, solvent was evaporated under reduced pressure to give the product as a red solid. This product was directly used for next reaction without purification assuming quantitative yield.

9,10-bis((4-(3-(4-(1,2,2-

triphenylvinyl)phenoxy)propoxy)phenyl)ethynyl)anthracene (DADT)



A mixture of 2-(4-(3-(4-iodophenoxy)propoxy)phenyl)ethane-1,1,2-triyl)tribenzene **6** (180 mg, 2.95 mmol), 9,10-diethynyl anthracene **9** (30.42 mg, 1.35 mmol) and $PdCl_2(PPh_3)_2$ (10 mg, 13.45 mmol), triethylamine (6 mL) was taken in a two necked flask. In another small flask CuI (5.12 mg, 26.89 mmol) was suspended in dry THF. Both the flasks were degassed separately by repeated freeze-pump-thaw cycles. After this, CuI was mixed with the reaction mixture and kept for reflux at 70 °C under argon atmosphere for 24 hours. The progress of the reaction was monitored by TLC (ethyl acetate/hexane, 2:18). After the completion of the reaction, the crude mixture was filtered through celite and washed with DCM. After removal

of DCM *in vacuo*, the residue was purified by silica column chromatography using hexane/ ethyl acetate and as eluent, affording the products as a red solid. Yield: 10%.

FTIR (v_{max} in cm⁻¹): 3021, 2927, 2362, 2401, 1601, 1215, 768. ¹H NMR (400 M Hz, CDCl₃), δ (TMS, ppm): 8.70 (m, 4H), 7.69 (d, J = 8.51, 4H), 7.66 (d, 4H) 7.09 (m, 34H), 6.70 (m, J = 8.07, 8H), 4.22 (td, 8H), 2.28 (m, 4H). MALDI-TOF-MS: [M⁺] calcd. for C₈₈H₆₆O₄: 1187.50, found: 1187.54.



9,10-bis((4-(3-(4-(1,2,2-triphenylvinyl)phenoxy)propoxy)phenyl)ethynyl)anthracene (DADT)

¹H NMR (top) and MALDI-TOF spectrum (bottom) spectra of 9,10-bis((4-(3-(4-(1,2,2 triphenylvinyl)phenoxy) propoxy)phenyl)ethynyl)anthracene.

Schemes, Tables and Figures

Scheme S3. Chemical structure of 1 and 2.

Scheme S4. Chemical structure of various second acceptors.

Scheme S5. Chemical structure of chiral-PBI based second acceptors.

Figure S1. Photograph of the **DADT** solutions under a) visible and b) UV (365 nm) lights in various solvents. 1. *o*-DCB, 2. *n*-Decane, 3. DMSO, 4. DMF, 5. *n*-Hexane, 6. CHCl₃, 7. DCM, 8. THF.

Figure S2. a) Emission spectrum of **DADT** in DCM (C = 1 m M, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$); inset is a zoomed portion (340-460 nm) of the emission spectrum of **DADT** showing the weak emission peak of TPE in **DADT**. b) Spectral overlap between normalised emission spectrum of **1** thin film ($\lambda_{ex} = 330 \text{ nm}$) and absorption spectrum of **DADT** (350-600 nm) in DCM solution.

Figure S3. a) Absorption spectra of **1**, **2** and physical mixture **1**+**2** (2:1) in DCM solution (*C* = 0.5 m M, l = 1 mm. Emission spectra of **1**, **2** and physical mixture **1**+**2** (2:1) in b) DCM (*C* = 0.5 m M, l = 1 mm, $\lambda_{ex} = 320$ nm) and c) thin film ($\lambda_{ex} = 340$ nm).

Figure S4. Emission lifetime decay profile of **1**, **2**, **1**+**2** (2:1) and **DADT** in a) DCM solution (C = 1 mM, l = 1 mm, $\lambda_{ex} = 374 \text{ nm}$, $\lambda_{mon} = 523 \text{ nm}$) and b) thin film ($\lambda_{ex} = 374 \text{ nm}$, $\lambda_{mon} = 526 \text{ nm}$).

	Lifetime (ns)				
	1	2	1+2	DADT	
	$\lambda_{\rm mon}$ = 426 nm for	$\lambda_{\rm mon}$ = 550 nm for	$\lambda_{\rm mon}$ = 550 nm for	$\lambda_{\rm mon}$ = 523 nm for	
	solution and 440 nm for	solution and 560 nm for	solution and 560 nm for	solution and 526 nm for	
	thin film	thin film	thin film	thin film	
Solution	3.30 ns (100%)	3.32 ns (100%)	0.12 ns (4.49%)	1.31 ns (12.66%)	
			1.39 ns (95.51%)	2.92 ns (87.34%)	
Thin film	0.95 ns (71.50%)	0.28 ns (08.47%)	0.35 ns (4.46%)	0.43 ns (18.70%)	
	1.60 ns (28.50%)	1.50 ns (38.64%)	2.22 ns (48.83%)	1.80 ns (51.79%)	
		4.10 ns (52.89%)	3.52 ns (46.71%)	8.60 ns (29.51%)	

Table S1. Comparison of the emission lifetime of 1, 2, 1+2 (2:1) and **DADT** ($\lambda_{ex} = 374 \text{ nm}$).

Figure S5. Comparison of the absorption and emission spectra of the **DADT** ($\lambda_{ex} = 320$ nm) with a) RB and b) PBI-1, c) DPP, d) NR, e) RBn in DCM solution ($\lambda_{ex} = 553$ nm for RB, 488 nm for PBI-1, 538 nm for NR, 512 nm for DPP, 493 nm for RBn and 320 for DADT).

Figure S6. Comparison of the spectral overlap between **DADT** emission ($\lambda_{ex} = 320$ nm) and absorption of a) RB, b) PBI-1, c) DPP, d) NR, e) RBn in DCM solution. Inset shows the corresponding spectral overlap integral value.

Figure S7. Normalized absorption spectrum of **DADT** thin film alone and containing 3-10 mol% of RB. As a representative example, **DADT**-RB thin film is used and similar observations were obtained for other acceptors also.

Table S2. Absolute fluorescence quantum yield (%) of **1**, **2**, **DADT**, **DADT** with 10 mol% of second acceptor RB.

Sample	Absolute fluorescence quantum yield (%)
1	64.76
2	7.11
DADT	1.96
DADT + 10 % of RB	1.60

Figure S8. Variation of the emission spectrum of **DADT** thin films upon addition of increasing amounts (0-10 mol%) of a) PBI-1, b) NR and c) RBn, ($\lambda_{ex} = 340$ nm).

Figure S9. Emission lifetime decay profile of **DADT** thin films with varying amounts (0-10 mol%) of a) PBI-1, b) NR and c) RBn ($\lambda_{ex} = 374$ nm, $\lambda_{mon} = 526$ nm). Insets show the fluorescence lifetime decay profile of the acceptors in the absence (**O**) and presence (Δ) of **DADT**, monitored at 580 nm for PBI-1 and 606 nm for NR and 600 nm for RBn ($\lambda_{ex} = 374$ nm).

NR alone is not emissive in thin film, but in the presence of **DADT** due to efficient energy transfer emission becomes active.

	Lifetime (ns)					
	DADT	DADT-DPP	DADT-RB	DADT-PBI-1	DADT-NR	DADT-RBn
	$(\lambda_{\rm mon} = 526 \text{ nm})$	$(\lambda_{\rm mon} = 560 \text{ nm})$	$\lambda_{\rm mon} = 582 \text{ nm}$)	$(\lambda_{\rm mon} = 580 \text{ nm})$	$(\lambda_{\rm mon} = 606 \text{ nm})$	$(\lambda_{\rm mon} = 600 \text{ nm})$
Solution	1.31 ns (12.66%)	0.32 ns (04.83 %)	0.90 ns (04.22 %)	0.36 ns (04.97 %)	0.32 ns (04.36 %)	0.33 ns (03.53 %)
	2.92 ns (87.34%)	2.40 ns (64.24 %)	2.18 ns (44.20 %)	2.27 ns (48.23 %)	2.36 ns (60.79 %)	2.37 ns (47.87 %)
		3.87 ns (30.91 %)	3.48 ns (51.58 %)	3.50 ns (46.80 %)	3.80 ns (34.86 %)	3.95 ns (48.60 %)
Thin film	0.43 ns (18.70%)	0.26 ns (3.55 %)	0.23 ns (3.52 %)	0.27 ns (3.30 %)	0.26 ns (3.40 %)	0.23 ns (2.36 %)
	1.80 ns (51.79%)	1.38 ns (12.51 %)	1.47 ns (15.25 %)	1.33 ns (12.58 %)	1.70 ns (21.41 %)	1.6 ns (17.62 %)
	8.60 ns (29.51%)	3.01 ns (83.95 %)	3.08 ns (81.52 %)	3.03 ns (84.12 %)	3.15 ns (75.19 %)	3.42 ns (80.01 %)

Table S3. Variation of the emission lifetime of **DADT** with 10 mol% of second acceptors $(\lambda_{ex} = 374 \text{ nm}).$

Figure S10. Variation of **DADT** emission with increasing amounts of a) DPP, b) RB, (0-20 mol%) in DCM (C = 1 mM, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$) and corresponding direct excitation ($\lambda_{ex} = 520 \text{ nm}$) of a) DPP and b) RB in presence of **DADT** (dotted lines). Comparison of the emission spectrum of **DADT**, second acceptors a) DPP, b) RB, (20 mol%) in the presence and absence of **DADT** (C = 1 mM, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$).

Figure S11. Variation of the absorption spectrum of **DADT** upon addition of increasing amounts (0-20 mol%) of a) DPP and b) RB in DCM (C = 1 mM, l = 1 mm).

Figure S12. Variation of the emission spectrum of **DADT** with increasing amounts of a) DPP, b) RB, c) PBI-1, d) NR, e) RBn (0-20 mol%) in DCM (C = 1 mM, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$).

Energy transfer studies are conducted in different concentrations of **DADT** in DCM (eg. $1x10^{-5}$ and $5x10^{-5}$ M) exhibited consistent results. Since our main focus is thin films of **DADT** prepared from 1 mM DCM solution, all energy transfer studies in solution state were conducted in the same concentration.

Figure S13. a)-b) Optical microscope images of the aggregates formed by DADT.

Figure S14. a)-c) TEM images of the aggregates formed by PBI-1.

Figure S15. a), b) Fluorescence microscopy images of **DADT**-PBI-1 sheets upon excitation at 352 nm and monitored at 573 nm.

Figure S16. a)-d) TEM images of DADT-PBI-1 nanosheets.

Figure S17. a)-c) SEM images of the **DADT**-PBI-1 nanosheets deposited on silicon wafer substrate and imaged without metal coating.

Figure S18. AFM images of the DADT-PBI-1 nanosheets with height profile.

Figure S19. AFM images of the DADT-PBI-1 nanosheets with height profile.

Figure S20. Schematic of the plausible pathways involved in the formation of DADT nanosheets.

Figure S21. Optical microscope images of the time dependent growth of DADT-PBI-1 nanosheets.

Figure S22. XRD profile of the DADT-PBI-1 and DADT-DPP nanosheets.

Figure S23. a)-c) TEM images of nanosheets aggregates formed by DADT-DPP.

Figure S24. Comparison of the absorption and emission spectra of the **DADT** with a) PBI-2R and b) PBI-2S ($\lambda_{ex} = 320$ nm for **DADT** and 490 nm for PBI-2R/S).

Figure S25. Spectral overlap integral between **DADT** emission and a) PBI-2R and b) PBI-2S absorption in DCM solution ($\lambda_{ex} = 320$ nm). Inset shows the corresponding spectral overlap integral.

Figure S26. Variation of the emission spectrum of **DADT** upon addition of increasing amounts of a) PBI-2R and b) PBI-2S (C = 1 mM, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$). The energy transfer efficiency is calculated to be 49% and 51% for PBI-2R and PBI-2S, respectively.

Energy transfer studies were repeated in different concentrations of **DADT** (eg. $1x10^{-4}$ and $1x10^{-5}$ M) exhibited consistent results.

Figure S27. Comparison of the emission spectra of a) PBI-2S and b) PBI-2R (20 mol%) in the presence ($\lambda_{ex} = 320$ nm) and in the absence of **DADT** (C = 1 mM, l = 1 mm, $\lambda_{ex} = 490$ nm).

Figure S28. Variation of the a) emission spectrum ($\lambda_{ex} = 340 \text{ nm}$) and corresponding b) emission lifetime decay profile of **DADT** thin film upon addition of increasing amounts (0-10 mol%) of PBI-2S monitored at 526 nm ($\lambda_{ex} = 374 \text{ nm}$). Inset shows the fluorescence lifetime-decay profile of PBI-2S in the absence (**O**) and presence (Δ) of **DADT** ($\lambda_{ex} = 374 \text{ nm}$, $\lambda_{mon} = 580 \text{ nm}$).

Figure S29. Variation of the a) emission spectrum ($\lambda_{ex} = 340 \text{ nm}$) and corresponding b) emission lifetime decay profile of **DADT** thin film upon addition of increasing amounts (0-10 mol%) of PBI-2R monitored at 526 nm ($\lambda_{ex} = 374 \text{ nm}$). Inset shows the fluorescence lifetime-decay profile of PBI-2R in the absence (**O**) and presence (Δ) of **DADT** ($\lambda_{ex} = 374 \text{ nm}$, $\lambda_{mon} = 580 \text{ nm}$).

	Lifetime (ns) ($\lambda_{ex} = 374$ nm)				
	DADT	DADT-PBI-2R	DADT-PBI-2S		
	$(\lambda_{\rm mon} = 526 \text{ nm})$	$(\lambda_{\rm mon} = 580 \text{ nm})$	$(\lambda_{\rm mon} = 580 \text{ nm})$		
Solution	1.31 ns (12.66%)	0.30 ns (02.20 %)	0.26 ns (03.85 %)		
	2.92 ns (87.34%)	2.47 ns (34.09 %)	2.63 ns (36.09 %)		
		3.25 ns (63.71 %)	3.35 ns (60.06 %)		
Thin film	0.43 ns (18.70%)	0.28 ns (02.02 %)	0.21 ns (03.26 %)		
	1.80 ns (51.79%)	2.23 ns (24.77 %)	1.56 ns (17.51 %)		
	8.60 ns (29.51%)	4.03 ns (73.21 %)	3.09 ns (79.23 %)		

Table S4. Variation of the emission lifetime of DADT with 10 mol% of PBI-2R and PBI-2S.

Figure S30. TEM images of the nanosheet aggregates formed by a), b) **DADT-**PBI-2S and c) **DADT-**PBI-2R.

Figure S31. XRD profile of the DADT-PBI-2R and DADT-PBI-2S nanosheets.

Figure S32. Schematic of the plausible pathways involved in the formation of chiral nanosheets of **DADT** in the presence of PBI-2S and PBI-2R.

Figure S33. Comparison of the variation of the emission intensity ratio of **DADT** at a) 492 nm and b) 535 nm with increasing amounts of PBI-1, PBI-2S and PBI-2R (C = 1 mM, l = 1 mm, $\lambda_{ex} = 320 \text{ nm}$).

In the case of chiral PBI derivatives, a reduced energy transfer is observed due to the geometrically restricted positioning of the acceptor units in the donor-acceptor assembly. Hence the energy transfer efficiency of chiral and achiral PBIs is compared by monitoring fluorescence intensity variations at 492 and 535 nm. It is clear that achiral PBI-1 exhibits improved energy transfer compared to the chiral ones.

References

S1. N. Song, D.-X. Chen, Y.-C. Qiu, X.-Y. Yang, B. Xu, W. Tian and Y.-W. Yang, *Chem. Commun.*, 2014, **50**, 8231-8234.

S2. L. Zhang, X. Lou, Y. Yu, J. Qin, and Z. Li, *Macromolecules*, 2011, 44, 5186-5193.

S3. W. Fudickar and T. Linker, Chem. Eur. J., 2011, 17, 13661-13664.