Enhancing Spin Coherence in Metallic Single Walled Carbon Nanotubes Utilizing Chiral Perturbations

Supporting Information

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1. Materials and Sample Preparation

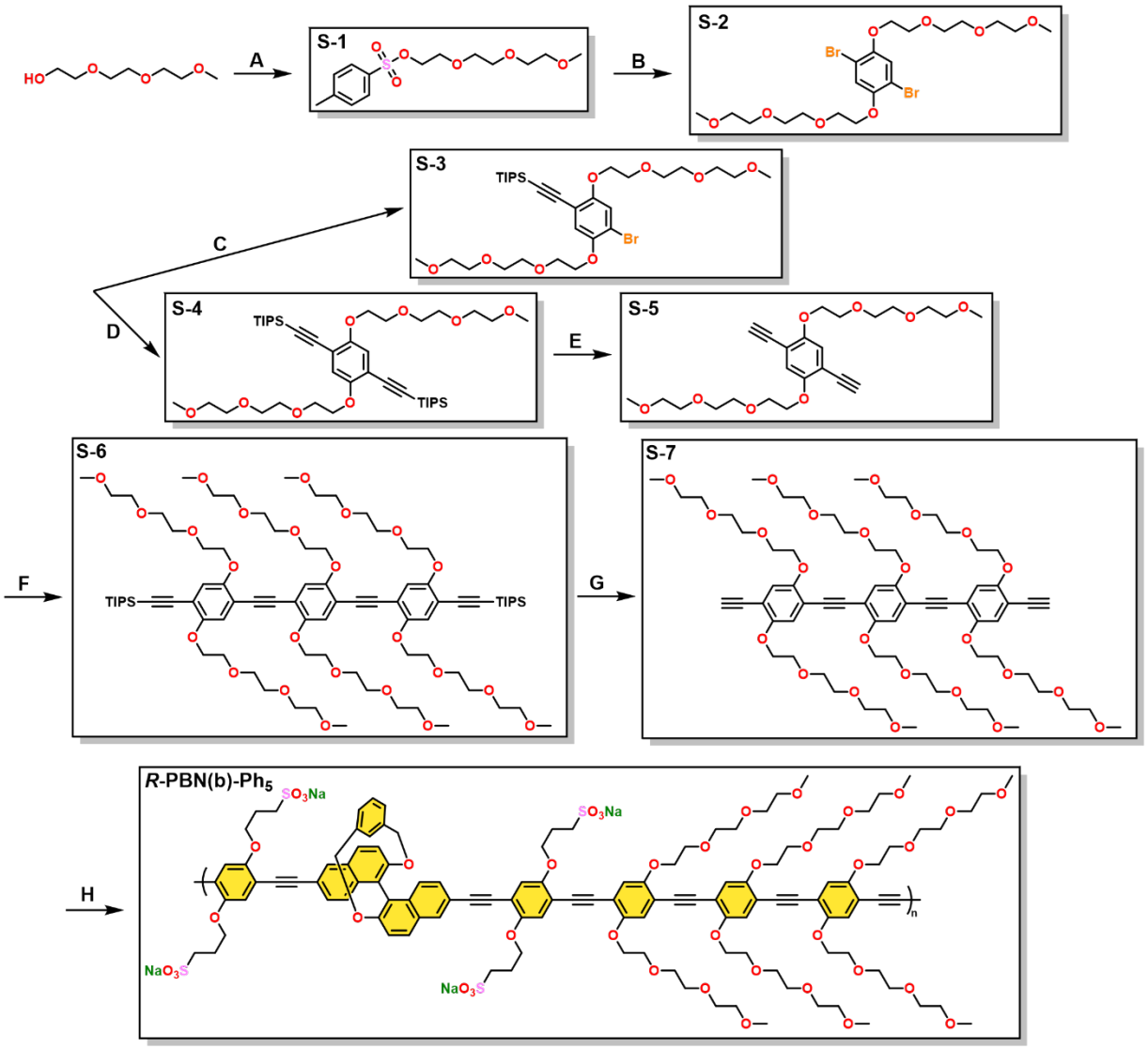
All manipulations were carried out under argon or nitrogen passed through an O_2 scrubbing tower (Schweitzerhall R3-11 catalyst) and a drying tower (Linde 3-Å molecular sieves) unless otherwise stated. Air sensitive solids were handled in a Braun 150-M glove box. Standard Schlenk techniques were employed to manipulate air-sensitive solutions. Tetrahydrofuran (THF), CH_2Cl_2, N,N-dimethylformamide (DMF), methanol, and diethyl ether were collected from a PURE SOLV (Innovative Technology) solvent purification system. All NMR solvents were used as received. All synthetic reagents and solvents were purchased from AK Scientific or MilliporeSigma and used as received without further purification. Sodium 3,3',3",3"'-((((4H,10H-5,9-(metheno)dinaphtho[2,1-b:1',2'-d][1,6]dioxacyclotridecine15,20-diyl)bis(ethyne-2,1-diyl))bis(5-bromobenzene-2,1,4- triyl))tetrakis(oxy))tetrakis(propane-1-sulfonate) was synthesized according to a previously published procedure[1]. Single-walled carbon nanotubes (SWNTs) were purchased from Nanointegris (IsoNanotubes-M-99% or IsoNanotubes-M-95%) and purified to obtain (11,11) nanotubes [see (11,11) Single Walled Carbon Nanotube Separation section]. Flash and size exclusion column chromatographic purification of non-ionic compounds was performed on the bench top, using respectively silica gel (EM Science, 230–400 mesh) and Bio-Rad Bio-Beads SX-1 as media.

1.1. (11,11) Single Walled Carbon Nanotube Separation

Single-walled carbon nanotubes (SWNTs) were purchased from Nanointegris (IsoNanotubes-M-99% or IsoNanotubes-M-95%) and purified to obtain (11,11) nanotubes using a modified version of a previously reported purification strategy.[1] In brief, the aqueous dispersed nanotubes from Nanointegris were filtered over a 100 kDa molecular weight cut off centrifugal filter (MilliporeSigma) and washed five times with water to remove the proprietary surfactant. The solid nanotubes were then redispersed in 5 mL of an aqueous solution of 1.04 % sodium deoxycholate (Sigma-Aldrich) at a concentration of 0.1 mg/mL via tip sonication for 2h at 0° C with a power of 20W. The nanotubes were then purified using a polyethylene glycol (6 kDa)/Dextran (70 kDa) aqueous two-phase extraction method. A centrifuge tube was charged with 4.8 mL of IsoNanotubes-M-99%/ IsoNanotubes-M-95% (0.1 mg/mL) in 1.04% DOC, 10 mL of 20% Dextran (70 kDa), 25 mL of 25% PEG (6 kDa), and 5 mL of H_2O. The centrifuge tube was then centrifuged for 3.5 min at 3,300 rpm. Then, 5 mL of the bottom phase was extracted and placed into a new centrifuge tube with 10 mL of 4% SC, and 10 mL 25% PEG (6 kDa). The centrifuge tube was then centrifuged for 3.5 min at 3,300 rpm. The light blue-green bottom phase containing single-electronic structure enriched (11,11) SWNTs was extracted from the centrifuge tube, filtered over a 100 kDa molecular weight cut off centrifugal filter (MilliporeSigma) and washed five times with water.

The achiral (11,11) SWNT sample was prepared by dispersing the purified (11,11) nanotubes in an aqueous solution of 0.55% sodium dodecyl sulfate (Sigma-Aldrich) via tip sonication for 2 h at 0 °C with a power of 20 W.

1.2. Molecular Synthesis



Scheme S1. Synthetic route to the *R*-PBN(b)-Ph_5 polymer. (**A**) 4-Toluenesulfonyl chloride in pyridine, 0 ºC to room temperature, overnight. (**B**) K_2CO_3 in DMF, 70 ºC, overnight. (**C**) TIPS acetylene, $Pd(PPh_3)_4$, CuI, and DIPA, in DMF, 60 ºC, overnight. (**D**) TIPS acetylene, $Pd(PPh_3)_4$, CuI, and DIPA, in DMF, 60 ºC, overnight. (**E**) TBAF in THF, 0 ºC to rt, 20 min. (**F**) $Pd(PPh_3)_4$, CuI, and DIPA, in DMF, 60 ºC, overnight. (**G**) TBAF in THF, 0 ºC to rt, 20 min. (**H**) Binol starting material[1], $Pd(PPh_3)_4$, CuI, and DIPA in DMF/H_2O 70 ºC, overnight.

1-(2-(2-(2-Methoxyethoxy)ethoxy)ethoxy)-4-(methylsulfonyl)benzene (S-1): 1-chloro-4-(methylsulfonyl)benzene (195 grams, 1.03 mol) was added to a round bottom flask containing 200 mL of pyridine and the flask cooled to 0 ºC. Triethylene glycol monomethyl ether (150 mL, 0.858 mol) was added to the flask and the reaction stirred overnight under argon. Afterwards, the mixture was diluted with 300 mL of water and extracted several times with methylene chloride. The organic layer was washed with 6M HCl (3x100 mL), sodium bicarbonate (3x100 mL) and brine (3x100 mL).The organic layers were combined, dried over Na_2SO_4, and filtered. The solvent was removed in vacuo to yield a light, pale yellow oil (191.4 g, 83%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.80 (d, 2H), 7.35 (d, 2H), 4.16 (t, 2H), 3.69 (t, 2H), 3.61 (m, 6H), 3.53 (m, 2H), 3.38 (s, 3H), 2.45 (s, 3H).

1,4-Dibromo-2,5-bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)benzene (S-2): 2,5-dibromobenzene-1,4-diol (30 g, 113.1 mmol) was placed in a round bottom flask with K_2CO_3 (25.8 g, 186.7 mmol). The system was evacuated by vacuum pump and subsequently filling with argon, 3x. 500 mL of dry DMF was sparged, cannulated into the round bottom, and the system heated to 70 ºC. **S-1** (89.4 g, 279.9 mmol) was added to the flask via a syringe. The solution was allowed to spin overnight at 80 ºC under argon. The crude mixture was then cooled to room temperature and extracted using ethyl acetate. The organic layers were combined and washed with brine. The collected organic layer was dried over Na_2SO_4, filtered, and the solvent removed in vacuo. The crude reaction material was further purified using a silica gel column with 4:3:3 EtOAc/hexanes/methylene chloride (25.1 g, 40%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.15 (s, 2H), 4.05 (t, 4H), 3.79 (t, 4H), 3.77 (t, 4H), 3.69 (t, 4H), 3.60 (m, 8H), 3.47 (t, 4H), 3.29 (s, 6H).

((4-Bromo-2,5-bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)phenyl)ethynyl) triisopropylsilane (S-3): To a round bottom flask was added **S-2** (4 g, 7.16 mmol), tetrakis(triphenylphosphine)palladium(0) (1.66 g, 1.43 mmol), and copper(I) iodide (136 mg, 0.716 mmol). The flask was evacuated by vacuum pump and refilled with argon 3x. A solution of THF (60 mL), diisopropyl amine (20 mL), and (triisopropylsilyl)acetylene (0.73 mL, 3.25 mmol) was degassed by freeze pump thaw with argon and cannulated into the reaction flask. The reaction was heated to 60 °C and stirred overnight. After cooling to rt, the mixture was filtered through celite and concentrated in vacuo. The resulting crude product was purified using a silica gel column with 4:3:3 EtOAc/hexanes/methylene chloride (1.55 g, 36%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.04 (s, 1 H), 6.94 (s, 1 H), 4.10 (m, 4H), 3.84 (t, 2H), 3.79 (t, 2H), 3.75 (t, 2H), 3.69 (t, 2H), 3.64-3.60 (m, 8H), 3.52-3.50 (m, 4H), 3.34 (s, 6H), 1.09 (s, 21H).

((2,5-Bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)-1,4-phenylene)bis(ethyne-2,1-diyl))bis(triisopropylsilane) (S-4): To a round bottom flask was added **S-2** (4 g, 7.16 mmol), tetrakis(triphenylphosphine)palladium(0) (1.66 g, 1.43 mmol), and copper(I) iodide (136 mg, 0.716 mmol). The flask was evacuated by vacuum pump and refilled with argon, 3x. A solution of THF (60 mL), diisopropylamine (20 mL), and (triisopropylsilyl)acetylene (7.3 mL, 32.5 mmol) was degassed by freeze pump thaw with argon and cannulated into the reaction flask. The reaction was heated to 60 °C and stirred overnight. After cooling to rt, the mixture was filtered through celite and concentrated in vacuo. The resulting crude product was purified using a silica gel column with 4:3:3 EtOAc/hexanes/methylene chloride, and the fluorescent blue product band was collected. The solvent was removed to yield a crystalline white powder. (2.93g, 54%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 6.87 (s, 2 H), 4.12-4.08 (m, 4H), 3.82 (t, 4H) 3.70 (t, 4H), 3.64-3.61 (m, 8H), 3.52 (t, 4H), 3.34 (s, 6H), 1.09 (s, 42H).

1,4-Diethynyl-2,5-bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)benzene (S-5): S-4 (600 mg, 0.786 mmol) was placed in a round bottom flask and dissolved in THF (50 mL). The solution was cooled to 0

°C and a solution of tetrabutylammonium fluoride in THF (1.6 mL, 1.6 mmol) was added dropwise. The mixture was stirred under an inert atmosphere for 20 minutes, warmed to room temperature, and stirred an additional 10 minutes. A mixture of methanol (10 mL) and water (0.1 mL) was added to quench the reaction. The mixture was reduced in vacuo and purified via column chromatography using a gradient of pure hexanes to 95:5 CH_2Cl_2/MeOH (353 mg, ~99% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.00 (s, 2H), 4.15 (t, *J* = 4.98 Hz, 4H), 3.87 (t, *J* = 4.93 Hz, 4H), 3.77 (dd, *J* = 6.00, 3.56 Hz, 4H), 3.66 (q, *J* = 5.17 Hz, 8H), 3.55 (dd, *J* = 5.66, 3.71 Hz, 4H), 3.38 (s, 6H), 3.33 (s, 2H).

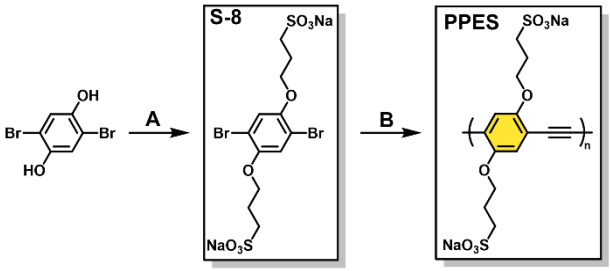
((((2,5-Bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)-1,4-phenylene)bis(ethyne-2,1-diyl))bis(2,5-bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)-4,1-phenylene))bis(ethyne-2,1-diyl))bis(triisopropylsilane) (S-6): To a round bottom flask was added tetrakis(triphenylphosphine)palladium(0) (1.03g, 0.888 mmol), and copper (I) iodide (0.085 g, 0.444 mmol). The flask was evacuated by vacuum pump and refilled with argon 3x. THF (60 mL), DIPA (20 mL), **S-3** (3.23g, 4.88 mmol), and **S-5** (1g, 2.22 mmol) are combined in a RBF and sparged for 30 minutes. The solvent solution is cannulated into the reaction flask, and the reaction was heated to 60 ºC and stirred overnight. After cooling to rt, the mixture was filtered through celite and concentrated in vacuo. The crude product was purified using a silica gel with 97.5% 4:3:3 EtOAc/hexanes/methylene chloride mixture and 2.5% MeOH to elute fluorescent impurity. The mobile phase is then switched to 96% 4:3:3 EtOAc/hexanes/methylene chloride mixture and 4% MeOH to elute fluorescent product. The pure product is a dark viscous oil (1.61g, 45%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.03 (s, 2H), δ 6.99 (s, 2H), δ 6.96 (s, 2H), δ 4.20 (t, 8H), δ 4.14 (t, 4H), δ 3.83-3.90 (m, 12H), δ 3.73 -3.77 (m, 12H), δ 3.58-3.67 (m, 28H), δ 3.47-3.55 (m, 16H), δ 3.37 (s, 6H), δ 3.34 (s, 6H), δ 3.33 (s, 6H), δ 1.14 (s, 42H).

5,5'-((2,5-Bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)-1,4-phenylene)bis(ethyne-2,1-diyl))bis(2-ethynyl-1,4-bis(2-(2-(2-methoxyethoxy)ethoxy)ethoxy)benzene) (S-7): S-6 (600 mg, 372.16 µmol) was dissolved in 50 mL of THF and sparged in an RBF for 30 min at 0 ºC. Tetra-n-butylammonium fluoride (1.17 ml of 1M in THF, 1.17 mmol) was added dropwise. The mixture was stirred at 0 ºC for 20 minutes and 10 minutes at room temperature. A mixture of 40 mL: 0.4 mL MeOH/H_2O was added to the reaction. The reaction was extracted with methylene chloride and brine. The collected organic layers were dried over Na_2SO_4, filtered, and solvent removed in vacuo. The crude product was purified using a silica gel column in 95:5 methylene chloride/MeOH and another column with 2:1 acetone/hexanes to elute impurities then the mobile phase was switched to acetone to elute the fluorescent product (396 mg, 83%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.04 (s, 2H), δ 7.03 (s, 2H), δ 7.00 (s, 2H), δ 4.16-4.21 (m, 12H), δ 3.88 (t, 12), δ 3.77-3.80 (m, 12H), δ 3.58-3.69 (m, 24H), δ 3.47-3.55 (m, 12H), δ 3.37 (s, 6H), δ 3.35 (s, 6H), δ 3.34 (s, 2H), δ 3.33 (s, 6H).

***R*-PBN(b)-Ph$_5$:** To a microwave vial, **S-7** (396 mg, 304.73 µmol) and the *R*-enantiomer of sodium 3,3',3",3'"-((((4H,10H-5,9-(metheno)dinaphtho[2,1-b:1',2'-d][1,6]dioxacyclotridecine15,20-diyl)bis(ethyne-2,1-diyl))bis(5-bromobenzene-2,1,4- triyl))tetrakis(oxy))tetrakis(propane-1-sulfonate) (synthesized according to a previous literature procedure[1]) (422.65 mg, 304.73 µmol) were added. The microwave vial is taken into the glovebox where tetrakis(triphenylphosphine)palladium(0) (70.4 mg, 60.946 µmol) and copper (I) iodide (5.8 mg, 30.473 µmol) are added. The vial is sealed in the glovebox. A solution of DMF (9 mL), water (6 mL) and diisopropylamine (3 mL) was degassed by gently bubbling argon through the solution for 1.5 hours. The solvent was cannulated into the microwave vial and heated by the microwave at 160 °C for 1 hour. The mixture was then heated by an oil bath at 70 °C overnight. A

solution of K_2CO_3 (300 mg) and NaCl (300 mg) in 5 mL of water was added, and then the reaction was allowed to cool to rt. The mixture was added to a 700 mL solution of acetone: diethyl ether:methanol (5:4:1) and stirred for 30 minutes. The mixture was filtered, and the filtrate was dissolved in a 30 mL solution of 7:3 H_2O/MeOH. The mixture was then filtered through a cotton plug. The crude product (120 mg, 15% yield) was filtered using a 10 kDa molecular weight cut off filter, washed with water (x5), resuspended in 7:3 H_2O/MeOH, and purified using gel permeation chromatography on a preparative column (160x16 mm) loaded with the sephacryl-based separatory medium S-100 [Sigma Aldrich; MW fractionation range 1-100 kDa (dextran)]. Fractions were collected such that the polymerization length used to wrap nanotubes was 10-20 mer (**Figure S9**).

***R*-PBN(b)-Ph$_5$-[(11,11) SWNTs]:** The polymer-wrapped ***R*-PBN(b)-Ph$_5$-[(11,11) SWNT]** superstructure was obtained by self-assembly in aqueous solution. ***R*-PBN(b)-Ph$_5$** polymer (OD = 2.6, 1cm cuvette) and 3:7 MeOH/H_2O (4.8 mL) were placed in a small vial with a septum cap and a stir bar. Then, aqueous two-phase extraction purified (11,11) SWNTs (4 mL, OD = 1.8, 1cm cuvette, 1.04% deoxycholate in H_2O) was added to the solution over 16 hours via a syringe pump. This solution was allowed to stir overnight. The solution was then filtered and washed with water (x5) through a 100 kDa molecular weight cut off centrifugal filter. The remaining solid was resuspended in 2 mL of a buffer solution (5 mM carbonate buffer in 3:7 MeOH:H_2O). Free, unbound polymer was removed using gel permeation chromatography. The 2 mL mixture was injected into a series of two preparative columns (160x16 mm each) loaded with sephacryl-based separatory medium connected in the order of S-500 [Sigma Aldrich; MW fractionation range 40-20000 kDa (dextran) and S-200 (Sigma Aldrich; MW fractionation range 1-80 kDa (dextran)], mounted on a GE/ÄKTA purifier HPLC system (GE Healthcare Bio-Science AB, Björkgatan, Uppsala, Sweden), and eluted with a 5 mM carbonate buffer in 3:7 MeOH:H_2O at a flow rate of 1 ml/min; three-wavelength detection (carbon nanotubes were detected at 650 nm; ***R*-PBN(b)-Ph$_5$** polymer was detected at 389 and 310 nm) was used to identify fractions that did not contain SWNTs. The fractions were collected as 1 ml aliquots; the polymer/SWNT fractions eluted at an earlier time (18-25 min range) followed by the free, unbound polymer (see Figure S**1**). Polymer/CNT containing fractions (eluting over an 18-25 min range) were collected and desalted via filtering over a 100 kDa molecular weight cut off centrifugal filter and washed with water five times. The remaining solid was resuspended in 3:7 MeOH:H_2O and filtered through a cotton plug before drop casting on devices.



Scheme S2. Synthetic route to the **PPES** polymer. (**A**) NaOH and 1,3-propane sultone in water/1,4-dioxane, room temperature, overnight. (**B**) 1,2-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)ethyne, $Pd(OAc)_2$/TXPTS (L/Pd = 3.4), and NaOH in water, 150 ºC, 90 min.

Sodium 3,3'-((2,5-dibromo-1,4-phenylene)bis(oxy))bis(propane-1-sulfonate) (S-8): 2,5-Dibromohydroquinone (10 g, 37 mmol), sodium hydroxide (3.73 g, 93.3 mmol), and 1,3-*p*ropane sultone (11.40 g, 93.3 mmol) was added to a round bottom flask containing 200 mL of water and 40 mL of 1,4-dioxane. This flask was stirred for 70 hours at room temperature. Then, the solution was poured into 350 mL acetone. The white solid was precipitate was collected via vacuum filtration, washed with acetone, and left to dry (18.15 g, 87% yield). ^{1}H NMR (400 MHz, DMSO-d6): δ 7.31 (s, 2H), 4.08 (t, 4H), 2.56 (m, 4H), 1.97 (m, 4H).

Poly[p-{2,5-bis(3-propoxysulfonicacid sodium salt)}phenylene]ethynylene (PPES): A 5 mL microwave vial (Biotage) was charged with **S-8** (0.167 g, 0.300 mmol), 1,2-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)ethyne (0.084 g, 0.302 mmol), Palladium(II) acetate (0.001 g, 0.005 mmol), tris(4,6)-dimethyl-3-sulfonatophenyl)phosphine trisodium salt (TXPTS) (0.010 g, 0.015 mmol) and a magnetic stir bar. The vial was then sealed with a vial cap, subjected to vacuum and charged with nitrogen (x3). An aqueous sodium hydroxide (5 mL, 1.5 M) solution was sparged with nitrogen for 1 hour and then added to the microwave vial via an air free syringe transfer. The vessel was then placed into a microwave cavity and heated to 150 °C for 90 minutes. The crude product was filtered using a 10 kDa molecular weight cut off filter, washed with water (x5), resuspended in 7:3 H_2O/MeOH, and purified using gel permeation chromatography on a preparative column (160x16 mm) loaded with the sephacryl-based separatory medium S-100 [Sigma Aldrich;MW fractionation range 1-100 kDa (dextran)]. Fractions were collected such that the polymerization length used to wrap nanotubes was 15-35 mer (**Figure S10**).

PPES-[(11,11) SWNTs]: (11,11) SWNTs (2 mg) and 2.5 mL of PPES (1.35 mg/mL) were placed in a small vial and tip sonicated at 2 W (0.8 W/mL) for 30 minutes, vortexed, and then tip sonicated at 10 W (4 W/mL) for 30 minutes. This solution was then centrifuged at 35000 g for 3 hours. The upper 60% supernatant was collected as **PPES-[(11,11) SWNTs].**

1.3. Fabrication of electrodes

Gold (Au) electrodes were fabricated on Si/SiO_2 substrates (1 um oxide layer) using standard electron-beam lithography (EBL) followed by thermal evaporation. A 5 nm titanium (Ti) adhesion layer and a 50 nm Au layer were sequentially deposited under high vacuum ($< 5 \times 10^{-6}$ Torr) inside an inert-atmosphere glove box to prevent oxidation and surface contamination.

The electrodes were patterned in a multi-channel cross-finger configuration, consisting of interdigitated pairs separated by approximately 400 nm. This geometry allowed multiple measurement channels on a single substrate, providing better contact coverage for the drop-cast SWNTs.

Conductance between adjacent electrode pairs was measured prior to SWNT deposition to confirm complete electrical isolation of the cross-finger electrodes. This ensured that all subsequent conduction originated exclusively from the deposited SWNT networks or molecular assemblies.

2. Instrumentation

2.1. Microwave Chemical Reactor

Microwave-assisted reactions were performed using an Emrys Personal Chemistry System (Biotage).

2.2. High Performance Liquid Chromatography (HPLC)

Free, unbound polymer in each polymer/SWCNT sample was removed using a GE/ÄKTA purifier HPLC system (GE Healthcare Bio-Science AB, Björkgatan, Uppsala, Sweden) equipped with two preparative columns (160x16 mm each; stationary phase: sephacryl S-500 and S-200) connected in a series. The HPLC system uses three-wavelength detection, which distinguishes fractions that contain SWCNTs.

2.3. NMR - Nuclear Magnetic Resonance Spectroscopy

^1H NMR (400 MHz) spectra and were acquired at room temperature in $CDCl_3$ or DMSO-d_6 on an AC-Bruker spectrometer. Chemical shifts (δ) are reported in parts per million (ppm) using residual protonated solvent as an internal reference.

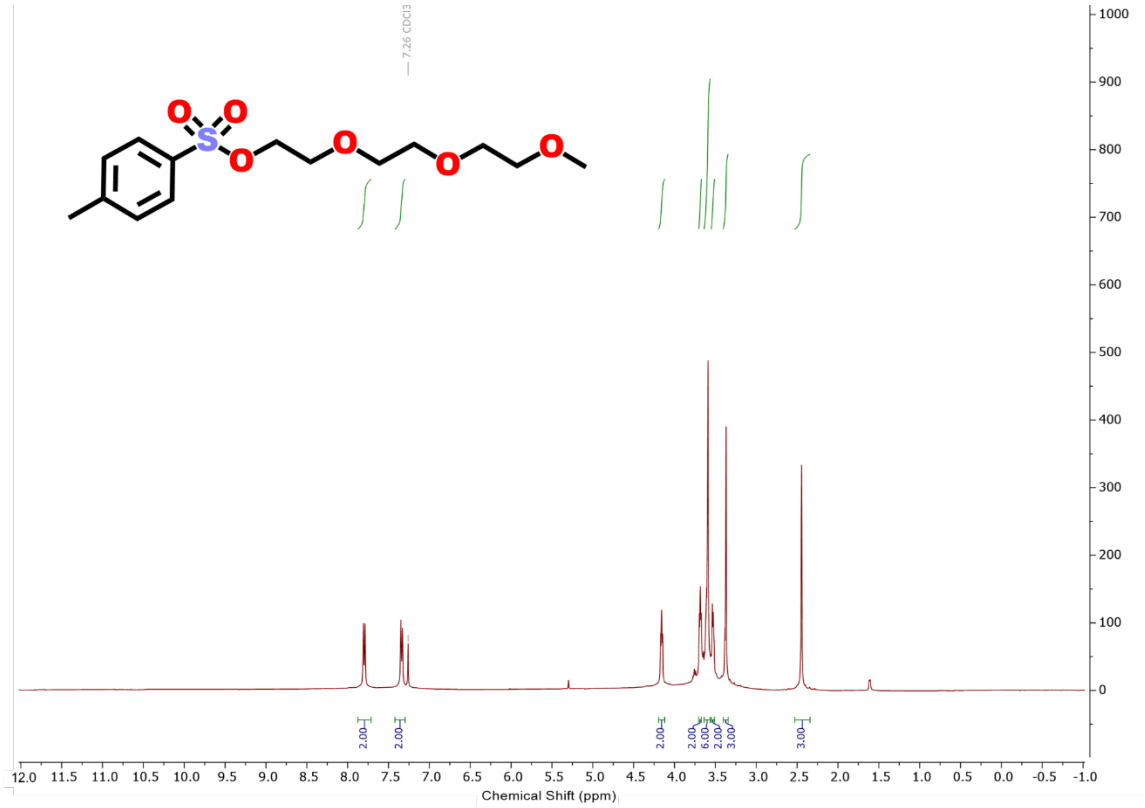


Figure S1. ^{1}H NMR (400 MHz) spectrum of **S-1** in $CDCl_3$ solvent at 25 ℃.

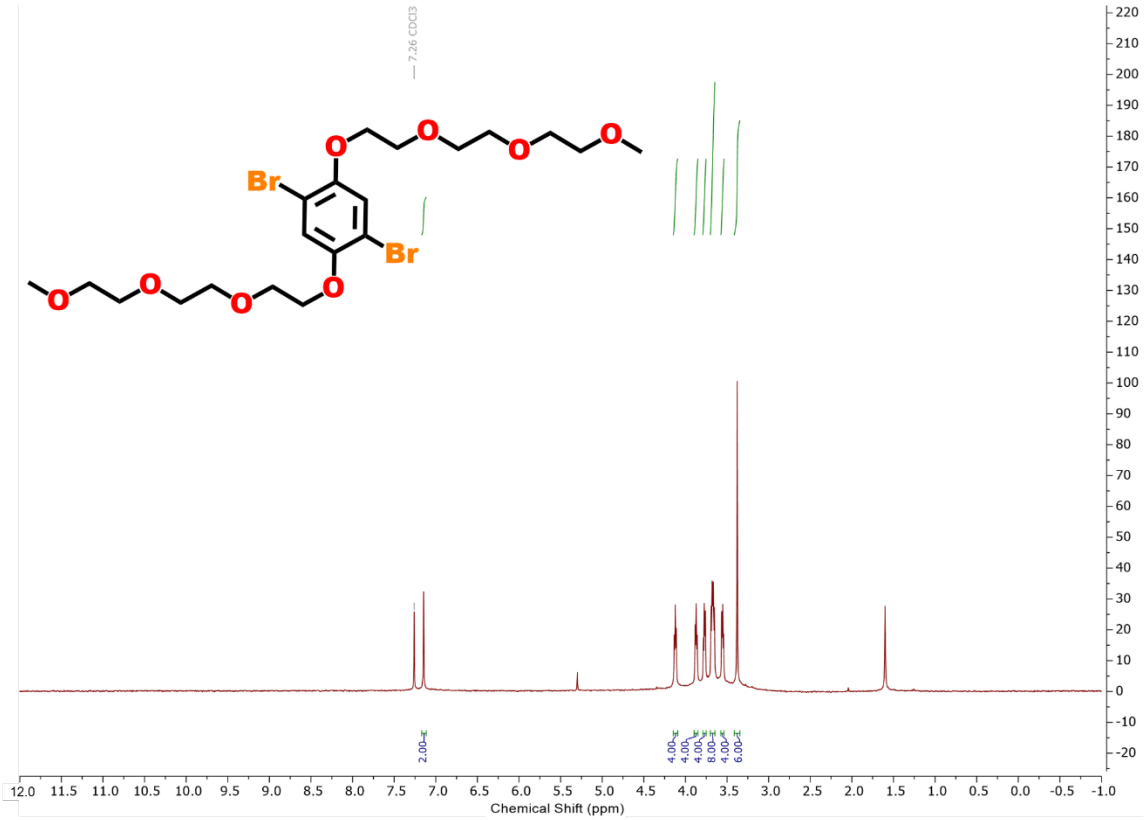


Figure S2. ^{1}H NMR (400 MHz) spectrum of **S-2** in $CDCl_3$ solvent at 25 ℃.

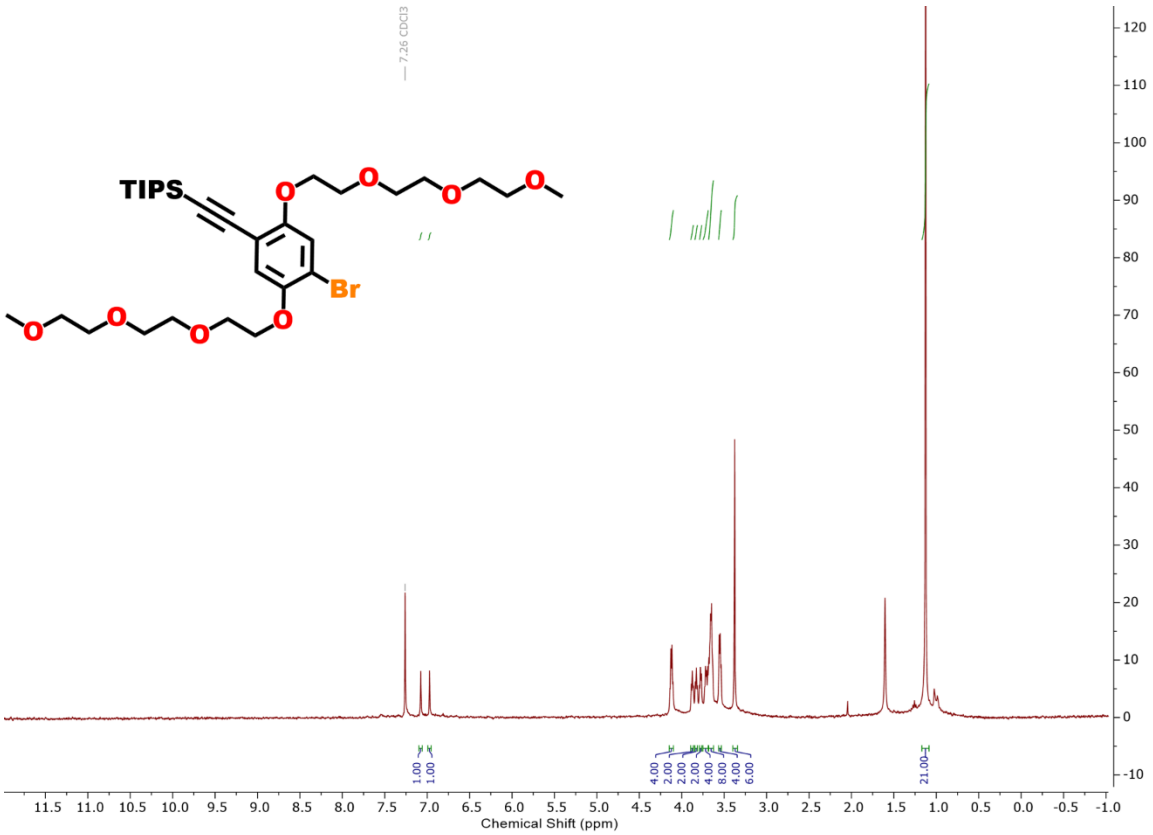


Figure S3. ^{1}H NMR (400 MHz) spectrum of **S-3** in $CDCl_3$ solvent at 25 °C.

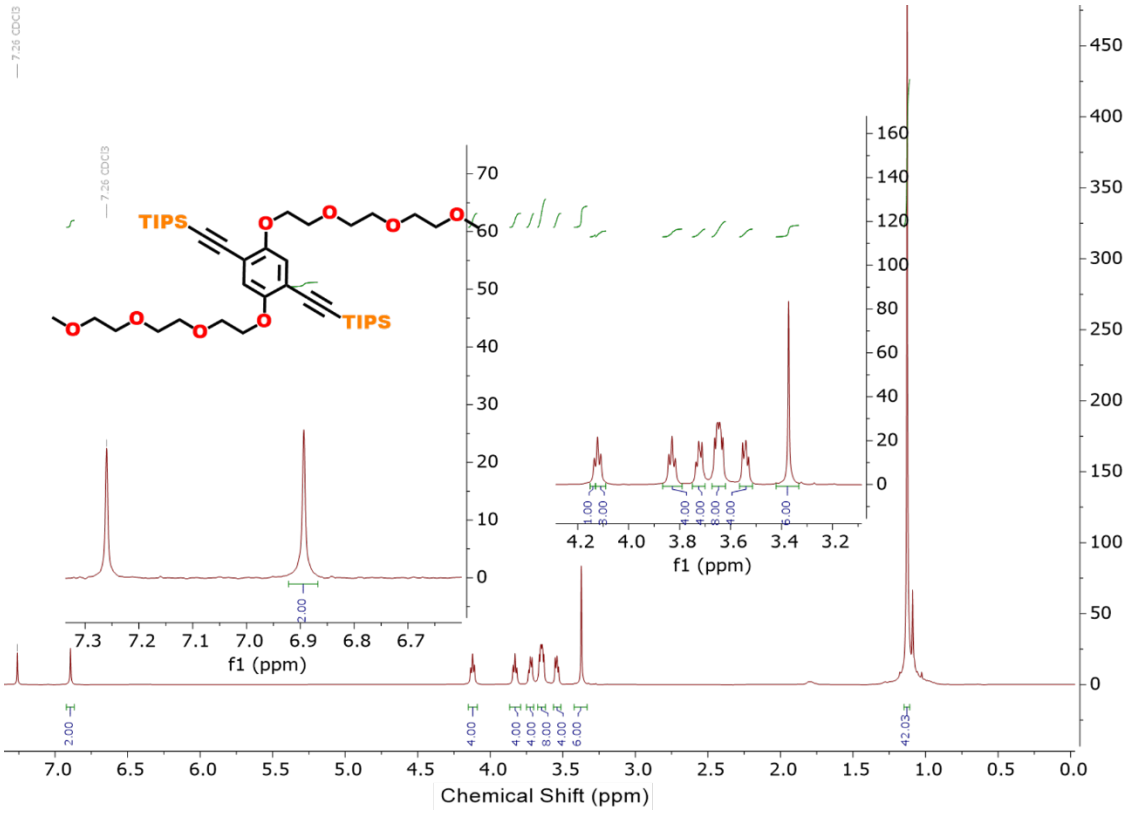


Figure S4. ^{1}H NMR (400 MHz) spectrum of **S-4** in $CDCl_3$ solvent at 25 °C.

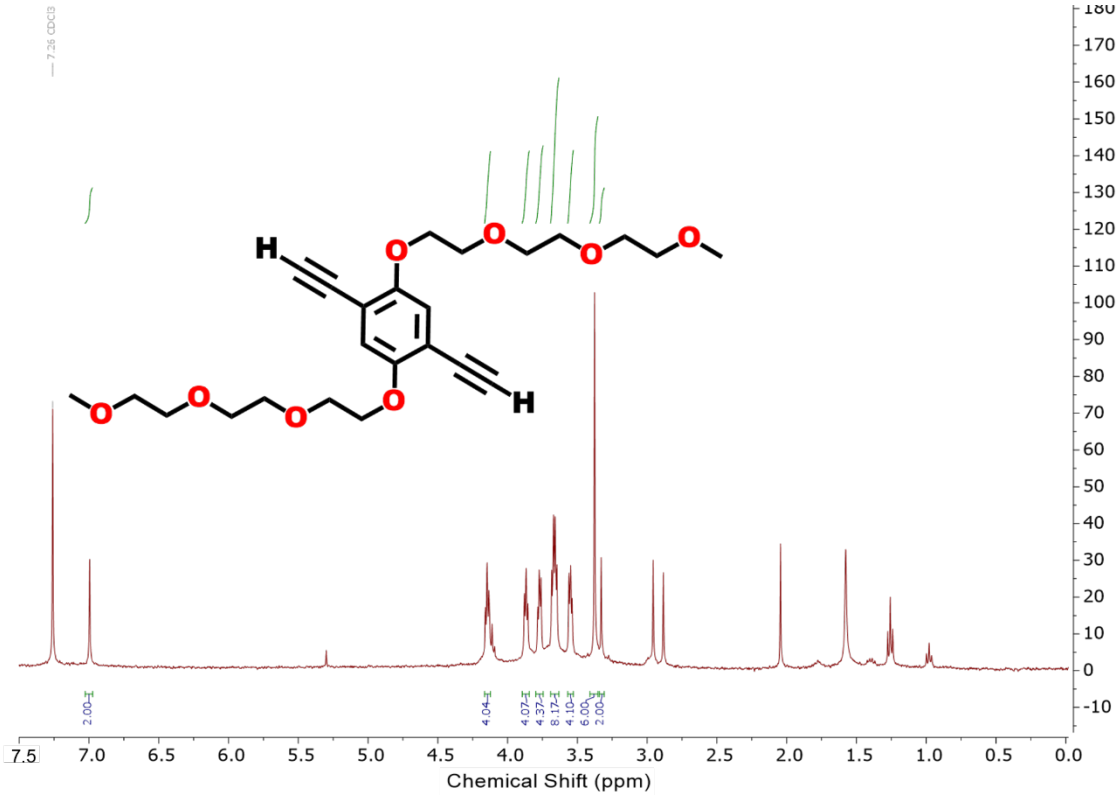


Figure S5. ^{1}H NMR (400 MHz) spectrum of **S-5** in $CDCl_3$ solvent at 25 °C.

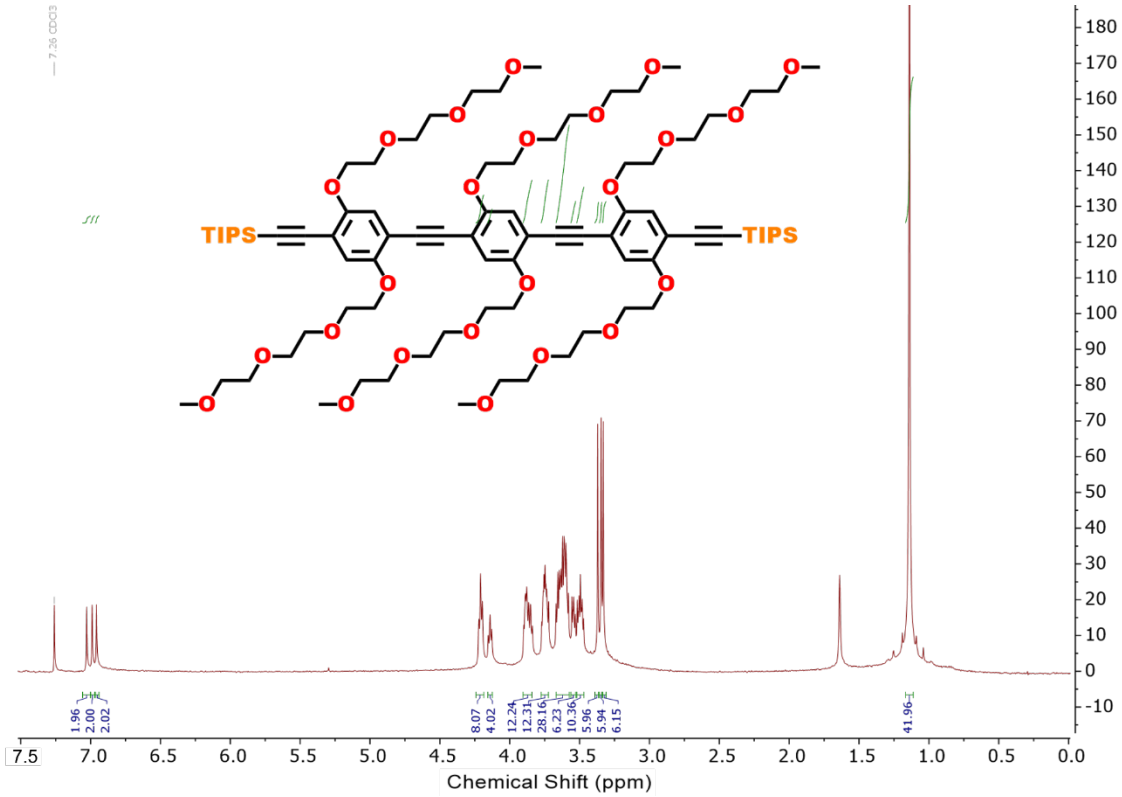


Figure S6. ^{1}H NMR (400 MHz) spectrum of **S-6** in $CDCl_3$ solvent at 25 °C.

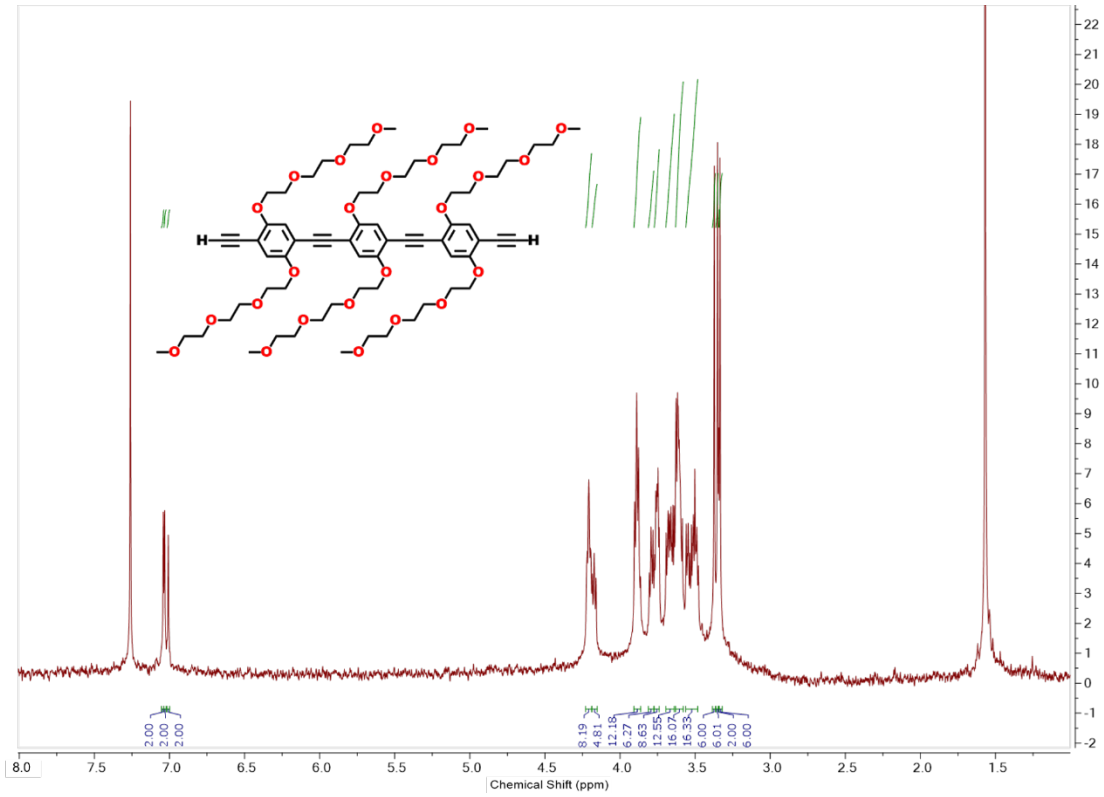


Figure S7. ^{1}H NMR (400 MHz) spectra of **S-7** in $CDCl_3$ solvent at 25 °C.

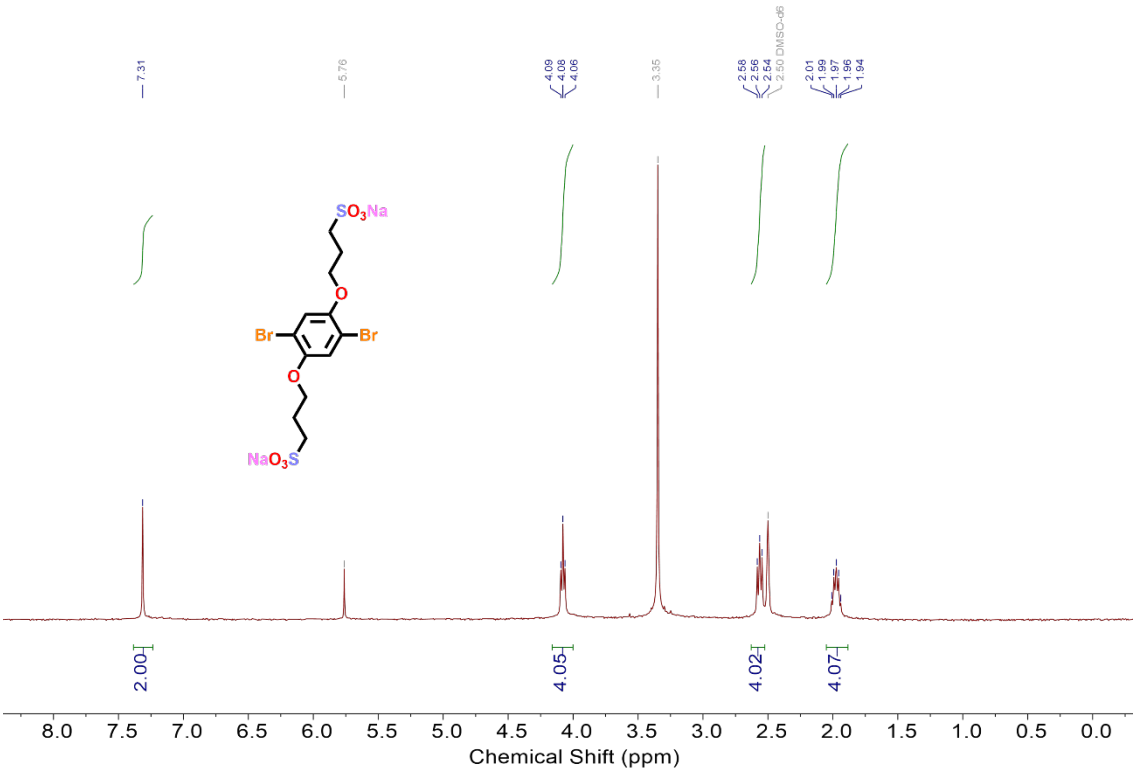


Figure S8. ^{1}H NMR (400 MHz) spectra of **S-8** in DMSO-d6 solvent at 25 ℃.

2.4. Gel Permeation Chromatography Characterization

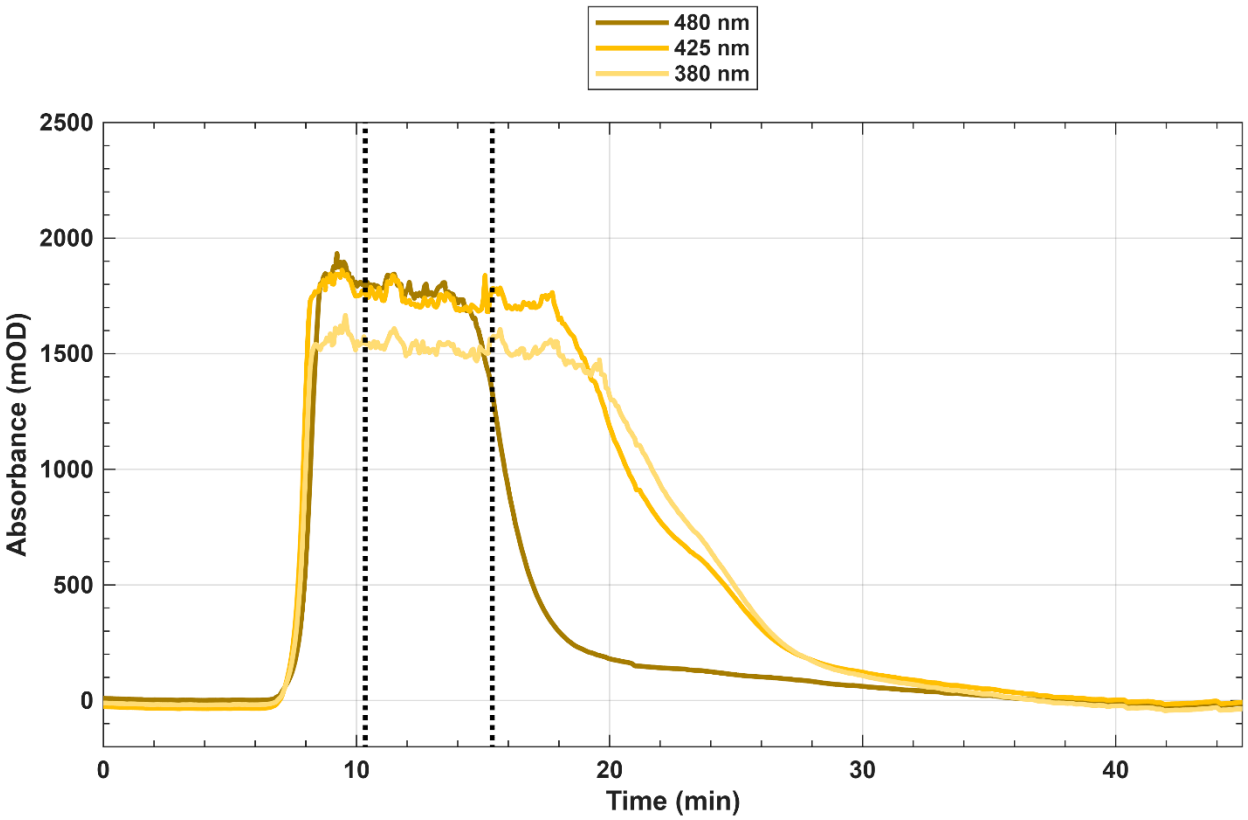


Figure S9. Gel Permeation Chromatogram of the ***R*-PBN(b)-Ph$_5$** polymer. The dotted black lines indicate the fractions collected and used to wrap SWCNTs; the degree of polymerization within this dotted black line elution time range is 10-20.

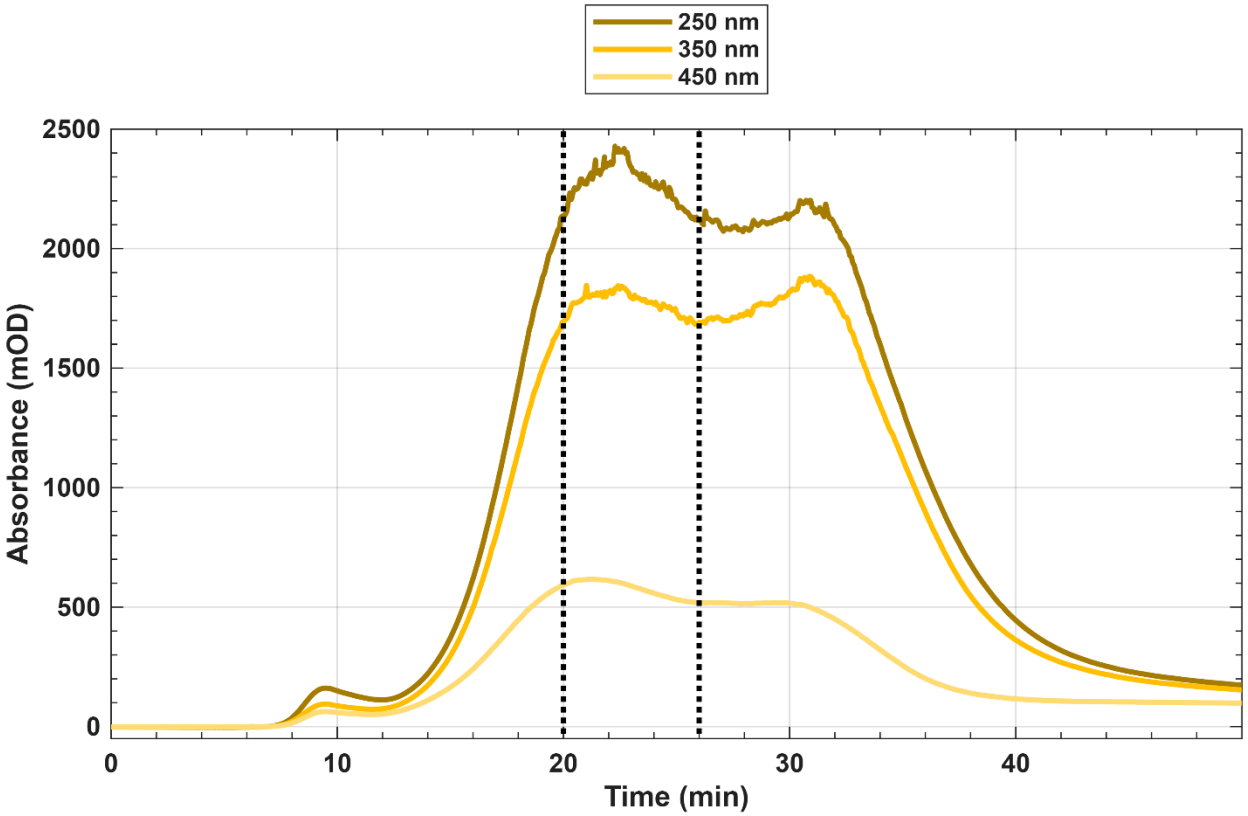


Figure S10. Gel Permeation Chromatogram of the **PPES** polymer. The dotted black lines indicate the fractions collected and used to wrap SWCNTs; the degree of polymerization within this dotted black line elution time range is 15-35 mer.

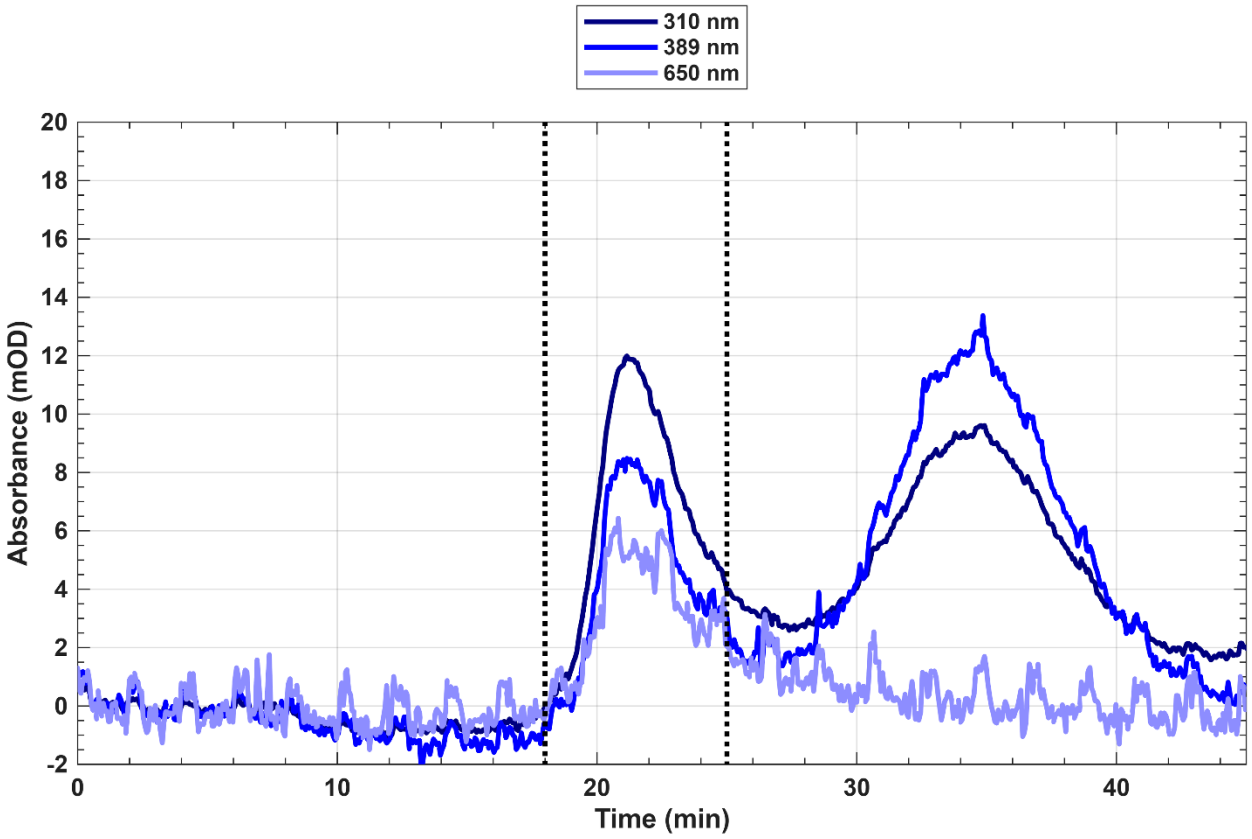


Figure S11. Gel Permeation Chromatogram of ***R*-PBN(b)-Ph$_5$-[(11,11) SWNTs]**. The dotted black lines indicate the fractions collected containing **R-PBN(b)-Ph$_5$-[(11,11) SWNTs]** without excess unbound ***R*-PBN(b)-Ph$_5$** polymer. Note that the 650 nm wavelength is resonant with an electronic transition of the (11,11) SWCNTs at which the polymer does not absorb.

2.5. Electronic Absorbance Spectroscopy

Electronic absorption spectra were taken on a Varian Cary 5000 UV-Vis-NIR Spectrophotometer in double beam mode with a blank solvent cuvette in the back sample holder. For all measurements, the spectral band width was set to 2 nm and the slit height was set to the "full" position. Data were collected every 1 nm with a scan rate of 600 nm/min. The data were baseline corrected to a baseline scan absent of the sample and blank (air).

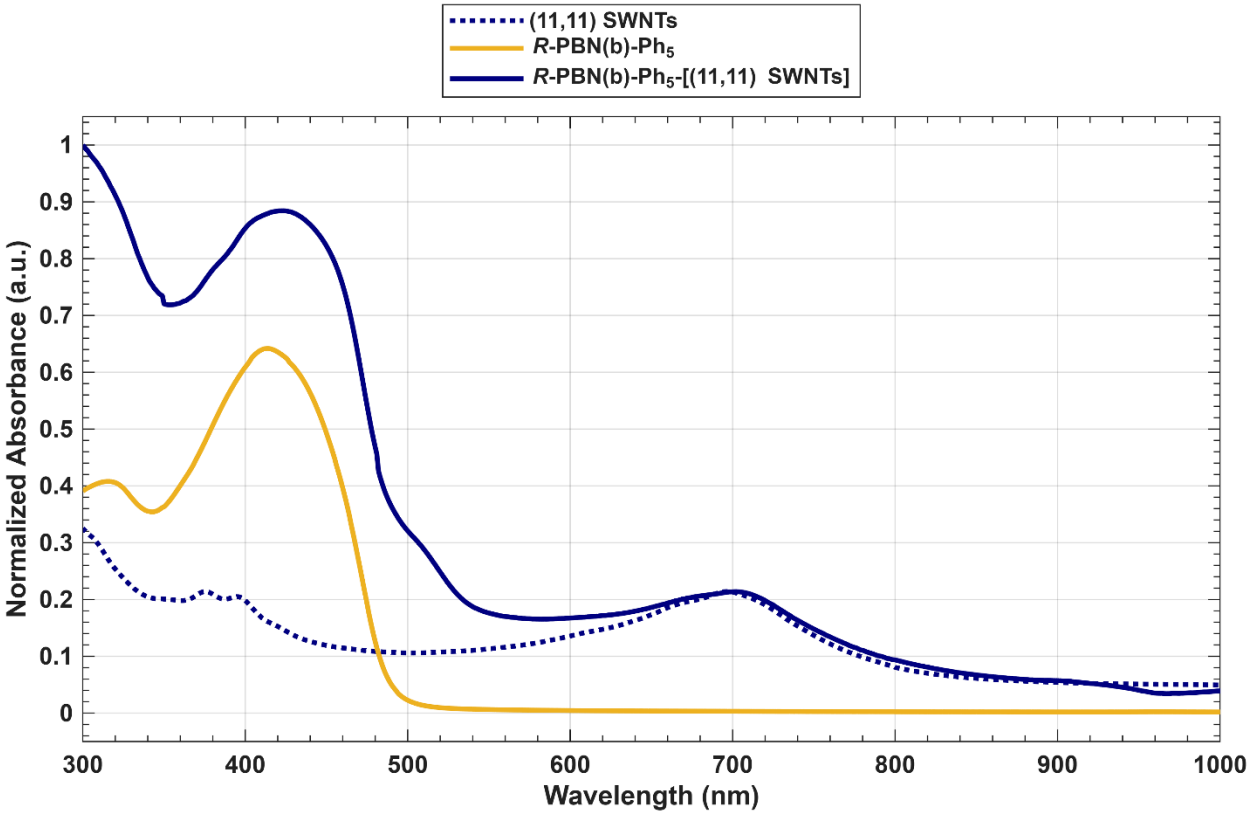


Figure S12. Electronic absorption spectra of the ***R*-PBN(b)-Ph$_5$-[(11,11)-SWNT]** superstructure, the (11,11) SWNTs dispersed in 1.04% sodium deoxycholate, and the ***R*-PBN(b)-Ph$_5$** polymer.

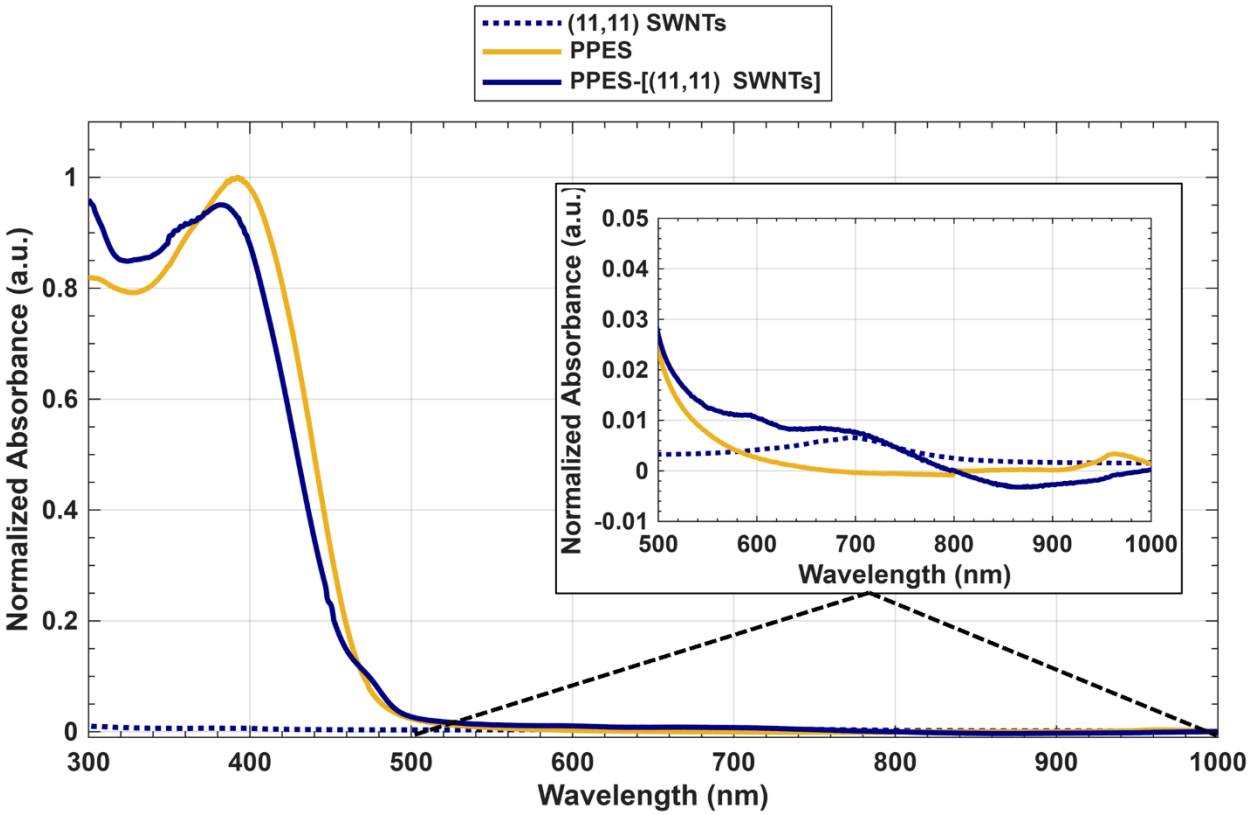


Figure S13. Electronic absorption spectra of the **PPES-[(11,11)-SWNT]** superstructure, the (11,11) SWNTs dispersed in 1.04% sodium deoxycholate, and the **PPES** polymer.

2.6. Raman Spectroscopy

Raman spectra were collected on a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 633 nm excitation laser.

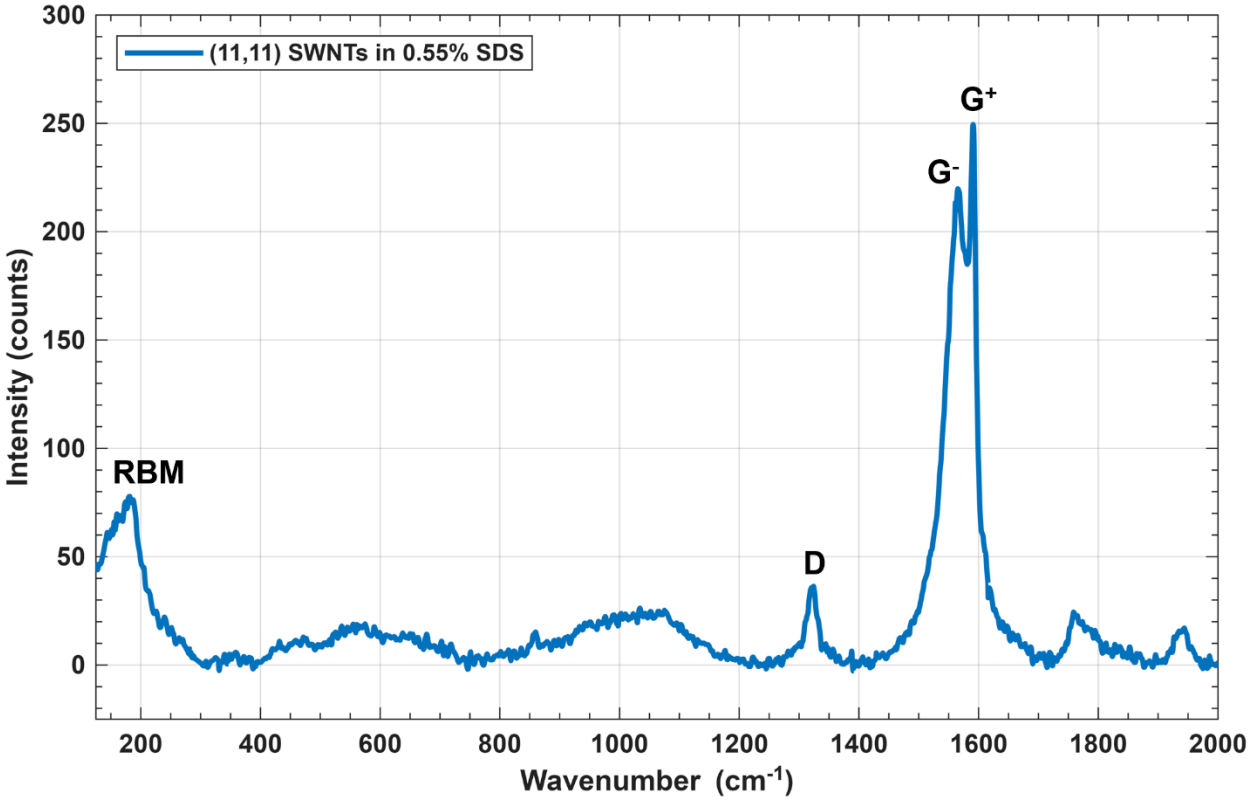


Figure S14. Raman spectrum of the **(11,11)-SWNTs** dispersed in 0.55% sodium dodecyl sulfate (SDS), 633 nm excitation. The prominent (11,11) Raman active vibrational modes are annotated: the radial breathing mode (RBM), the D band, the G^- band and the G^+ band.

3. Electrical / Magneto-Transport Measurements – Additional Results

3.1. I–V Characteristics of Chiral-Wrapped SWNT Devices

I–V characteristics of the ***R*-PBN(b)-Ph$_5$-[(11,11) SWNT]** chiral superstructures drop-cast onto Au electrodes are shown in **Figure S15**. The results exhibit ohmic-like behavior, similar to that observed for the chiral-on-top SWNT devices.

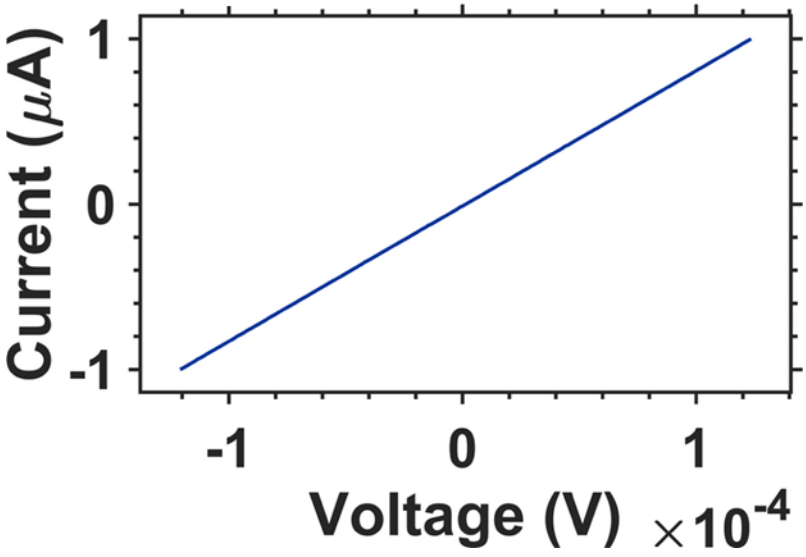


Figure S15. Electrical transport of a chiral-wrapped *R*-PBN(b)-Ph$_5$-[(11,11) SWNT] device. Current–voltage (I–V) characteristics recorded at 2 K, exhibiting ohmic behavior.

3.2. Temperature-Dependent Magnetoconductance

Temperature-dependent measurements for the device incorporating (11,11) SWNTs co-deposited with F-phenyl–L-α-helix polyalanine molecules are presented in **Figure S16.**

The WAL feature disappears upon increasing the temperature to 4 K.

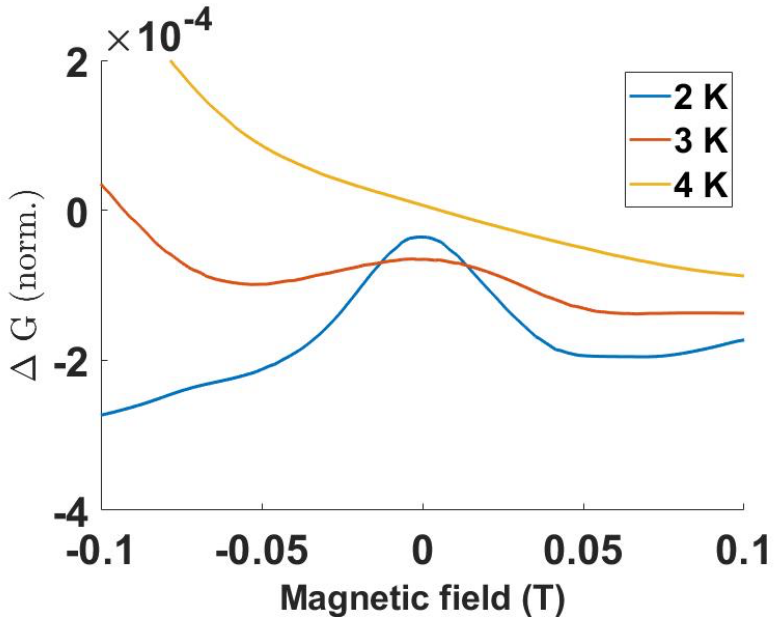


Figure S16. Temperature-dependent magnetoconductance of the (11,11) SWNT device co-deposited with F-phenyl–L-α-helix polyalanine molecules. *The weak antilocalization (WAL) feature gradually diminishes and disappears as the temperature increases to 4 K.*

Magnetoconductance measured at different temperatures for the (11,11) SWNT sample is presented in **Figure S17**. A clear weak localization (WL) feature is observed up to 30 K.

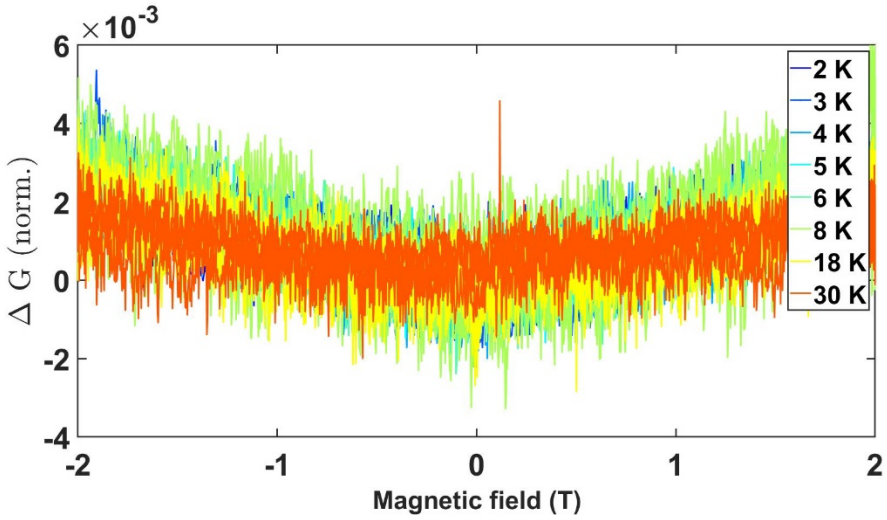


Figure S17. Magnetoconductance of the (11,11) SWNT *sample measured at different temperatures. Weak localization (WL) is clearly observed up to 30 K.*

Magnetoconductance in different temperatures for the achiral (11,11) SWNT sample is presented in **Figure S18.** Similar to devices that interrogate pristine (11,11) SWNTs, the achiral device exhibits weak localization (WL) that remains up to 30 K.

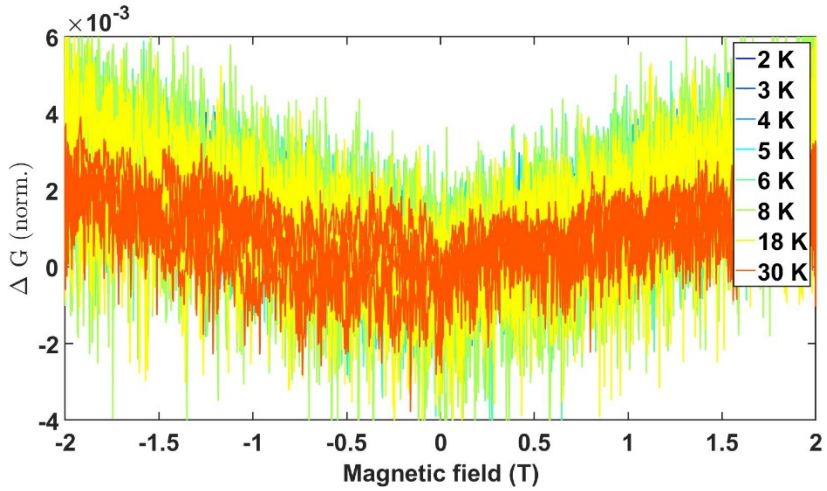


Figure S18. Magnetoconductance of (11,11) SWNTs co-deposited with achiral phenylacetic acid molecules measured at different temperatures. *Weak localization (WL) is clearly observed up to 30 K.*

3.3. Reference Measurements — Additional Results

Magnetoconductance measurements of the SWNTs uniformly wrapped in a racemic fashion by the achiral PPES polymer are presented in **Figure S19.**
The magneto-conductance does not exhibit either weak localization or weak antilocalization within the measurement resolution.

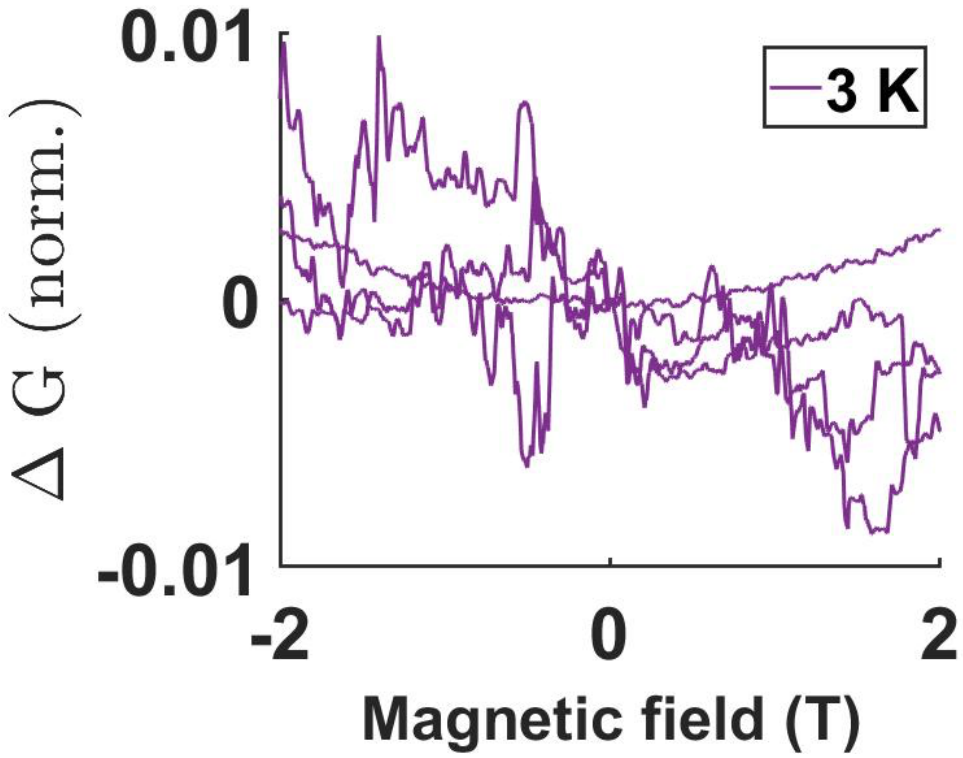


Figure S19. Magnetoconductance measurements of the SWNTs uniformly wrapped in a racemic fashion by the achiral PPES polymer. *A clear behaivor is not shown in this sample.*

For comparison, a reference measurement performed on the bare Au electrodes (without SWNTs bridging) showed a weak-localization-like response with virtually no indication of weak antilocalization (WAL) (**Figure S20**). This contrasts with the chiral-wrapped SWNT devices, which display a pronounced field-dependent modulation characteristic of strong spin–orbit–coupling-induced WAL. These observations confirm that the WAL originates from the chiral polymer–SWNT hybrid interface rather than from the metallic contacts.

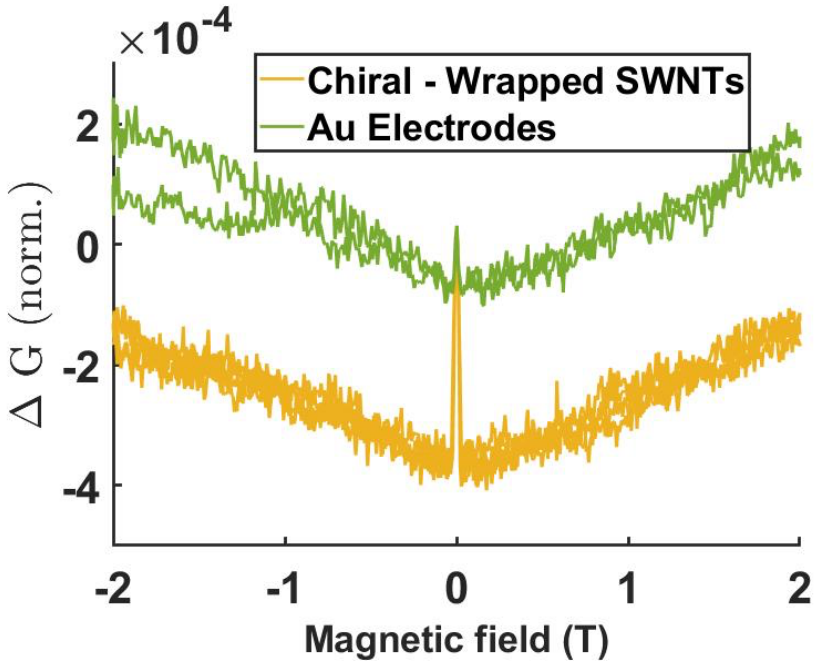


Figure S20. Comparison of magnetoconductance between the bare Au electrodes and the chiral-wrapped SWNT device. *The Au reference shows no WAL, while the chiral-wrapped device exhibits a clear WAL feature.*

4. Data Analysis and Fitting Procedures

4.1. Fitting model and fit example

The conductance G(B) was normalized according to:

$$\Delta G(B) = \frac{G(B) - G(0)}{G(0)}$$

This was done to highlight relative magnetoconductance changes independent of baseline resistance.

ΔG(B) was fitted to the **HLN** equation given in the main text **(Eq. 1),** extracting α, $L\varphi$, and $Lsoc$.

Figure S21 presents an example of the HLN fitting for a representative chiral-polymer wrapped SWNT device at 2 K. The measured magnetoconductance (dots) and the fitted curve (solid line) show excellent agreement in the low-field region, capturing the characteristic WAL cusp near zero magnetic field.

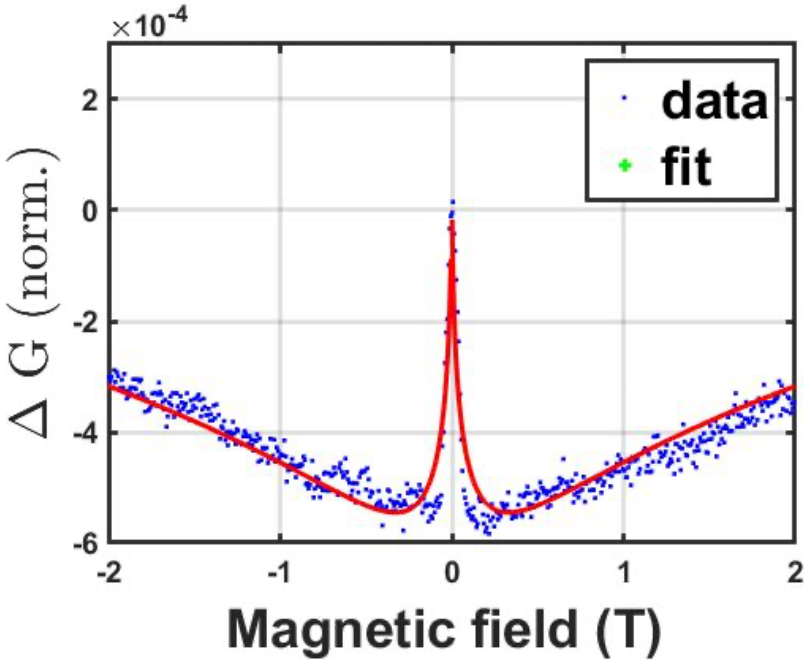


Figure S21. Example of HLN fitting for a chiral-polymer-wrapped SWNT device at 2 K. Experimental magnetoconductance data (dot) are fitted to the HLN model (solid line). The extracted parameters demonstrate the WAL-type behavior characteristic of chiral-induced spin–orbit coupling.

References

1. Mastrocinque, F. *et al.* Band gap opening of metallic single-walled carbon nanotubes via noncovalent symmetry breaking. *Proc. Natl. Acad. Sci.* **121**, e2317078121 (2024).