Supporting Information for:

A Single-molecule Potentiometer

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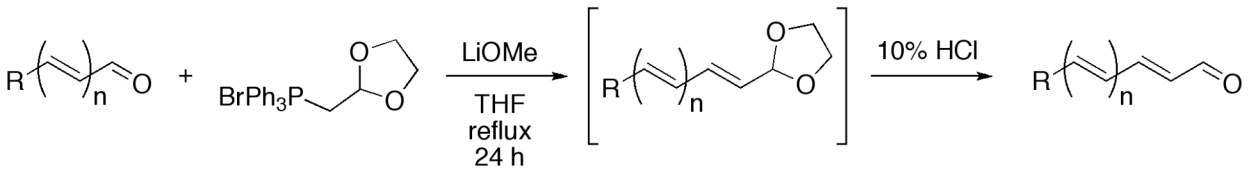
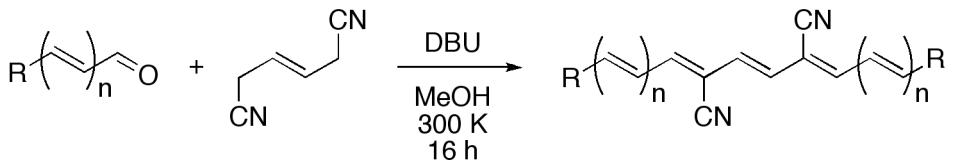
I. Synthetic Details:

General Information: (1,3-dioxolan-2-yl)methyl-triphenylphosphonium bromide was purchased from TCI America and stored in desiccator. The benzaldehydes, *trans*-cinnamaldehyde, 4-(methylthio)-benzaldehyde, 1,4-dicyano-2-butene, benzyl cyanide, 1,2-Epoxy-2-methylpropane, 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU) and all other reagents were ordered from Aldrich. All reactions were carried out under nitrogen unless otherwise noted. All chromatography was performed on a Teledyne ISCO Combiflash RF using Redisep RF silica columns. All 1H and 13C NMR were taken on a Bruker DRX300 (300MHz) unless otherwise noted. For more insoluble compounds a Bruker DRX400 (400MHz) and Bruker DMX500 (500MHz) were employed. Mass spec was found to be a poor way to characterize oligoene compounds.

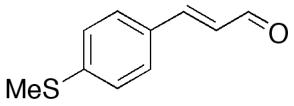


General Procedure for Wittig Homologation of Aldehydes: Lithium methoxide (2.6 eq.) in dry THF solution was added via syringe to a stirring solution of (1,3-dioxolan-2-yl)methyl-triphenylphosphonium bromide (2.5 eq.) in dry tetrahydrofuran (THF). The suspension was heated to reflux stirred for 30 minutes, changing color from off-white to light orange/pink. The aldehyde (1 eq.) in dry THF solution was added dropwise over 30-60 min. The suspension was refluxed for 24 h. The reaction suspension was then cooled to room temperature at which point 10% aqueous hydrochloric acid was added. Stirring was continued for 1 hour in order to hydrolyze the intermediate acetal, producing the all-*trans* enal. The organic layer was extracted with CH_2Cl_2 (x3) and the combined fractions were washed with water, sat. aqueous sodium bicarbonate solution and brine and dried over $MgSO_4$. Solvent was removed by rotary evaporation and the product was purified via column chromatography using an eluent gradient from 0% to 10% ethyl acetate in hexanes over 25 column volumes.

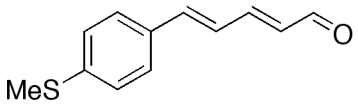


General Procedure for Double Knoevenagel Condensation: Aldehyde (150 mg) and 1,4-dicyano-2-butene (0.6 eq.) were dissolved in methanol or methanol/tetrahydrofuran mixture in a 100-mL round-bottomed flask. The flask was sealed with a rubber septum and purged with nitrogen for 10 minutes. 3.0 eq. of 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU) were added via syringe and the solution was left stirring for 16 hours. Upon completion of the reaction, the crude product was filtered off and washed with methanol and then purified by recrystallization in dichloromethane/methanol solution or column chromatography using dichloromethane/hexanes (1:1) as eluent.

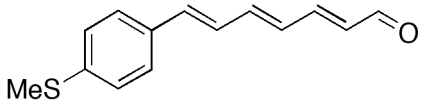
1-(4-Methylthiophenyl)-enals (R = MeS-C_6H_5):



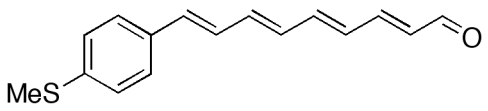
(*E*)-3-(4-(methylthio)phenyl)acrylaldehyde: Yield: 96%, as a pale yellow oil. 1H NMR (400MHz, $CDCl_3$): δ 9.70 (d, J = 7.6 Hz, 1H), 7.49 (d, J = 8.4 Hz, 2H), 7.45 (d, J = 16.0 Hz, 1H), 7.28 (d, J = 8.4 Hz, 2H), 6.69 (dd, J = 7.6 Hz, 15.6 Hz, 1H), 2.53 (s, 3H). 13C NMR ($CDCl_3$): δ 193.9, 152.2, 144.0, 130.8, 129.2, 128.0, 126.3, 15.4.



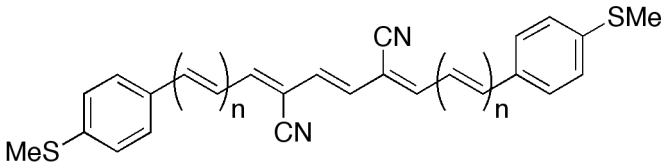
(2*E*,4*E*)-5-(4-(methylthio)phenyl)penta-2,4-dienal: Yield: 99%, as a yellow solid. 1H NMR ($CDCl_3$): δ 9.62 (d, J = 7.8 Hz, 1H), 7.43 (d, 2H), 7.26 (m, 3H), 6.95 (d, 2H), 6.27 (dd, J = 8.1 Hz, 15.0 Hz, 1H), 2.50 (s, 3H). 13C NMR ($CDCl_3$): δ 193.193.8, 152.1, 143.2, 140.3, 138.1, 131.4, 130.2, 127.7, 127.4, 126.8, 15.8.



(2*E*,4*E*,6*E*)-7-(4-(methylthio)phenyl)hepta-2,4,6-trienal: Yield: 73%, as an orange solid. 1H NMR ($CDCl_3$): δ 9.60 (d, J = 8.1 Hz, 1H), 7.38 (d, J = 8.4 Hz, 2H), 7.19 (m, 3H), 6.83 (m, 3H), 6.55 (dd, J = 11.1 Hz, 15.3 Hz, 1H), 6.19 (dd, J = 8.1 Hz, 15.3 Hz, 1H), 2.50 (s, 3H). 13C NMR ($CDCl_3$): δ 193.8, 152.1, 143.2, 138.2, 131.4, 130.2, 127.8, 127.4, 126.8, 15.8.



(2*E*,4*E*,6*E*,8*E*)-9-(4-(methylthio)phenyl)nona-2,4,6,8-tetraenal: Yield: 51%, as an orange-red solid. 1H NMR (400 MHz, $CDCl_3$): δ 9.58 (d, J = 8.0 Hz, 1H), 7.36 (d, J = 8.0 Hz, 2H), 7.19 (m, 3H), 6.85 (dd, J = 10.8 Hz, 15.2 Hz, 1H), 6.76 (dd, J = 11.2 Hz, 14.4 Hz, 1H), 6.64 (m, 2H), 6.18 (dd, J = 7.6 Hz, 15.2 Hz, 1H), 2.50 (s, 3H). 13C NMR (400 MHz, $CDCl_3$): δ 193.9, 152.2, 143.1, 139.4, 135.9, 134.6, 134.0, 132.0, 131.3, 130.3, 128.1, 127.5, 126.9, 16.0.



An; n = 0, 1, 2, 3, 4

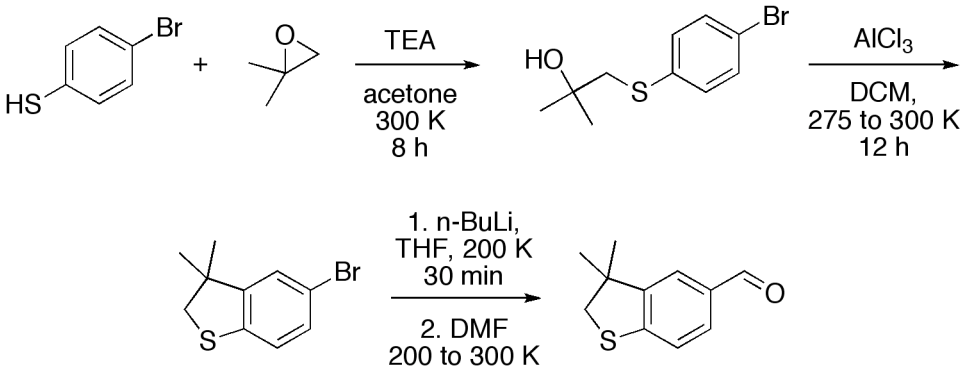
(2*Z*,3*E*,5*Z*)-2,5-bis(4-(methylthio)benzylidene)hex-3-enedinitrile (A0): Yield: 70%, as a lemon-yellow solid. 1H NMR ($CDCl_3$): δ 7.80 (d, J = 8.6 Hz, 4H), 7.30 (d, J = 8.0 Hz, 4H), 7.13 (s, 2H), 6.83 (s, 2H), 2.52 (s, 6H). 13C NMR (350K, $C_2D_2Cl_4$): δ 144.3, 143.3, 129.9, 129.8, 129.5, 126.1, 116.0, 108.5, 15.0.

(2*Z*,3*E*,5*Z*)-2,5-bis((*E*)-3-(4-(methylthio)phenyl)allylidene)hex-3-enedinitrile (A1): Yield: 45%, as a red solid. 1H NMR (CD_2Cl_2): δ 6.83 (d, J = 8.5 Hz, 4H), 6.64 (d, J = 8.4 Hz, 4H), 6.59 (dd, J = 114 Hz, 15.3 Hz, 2H), 6.38 (d, J = 11.4 Hz, 2H), 6.32 (d, J = 15.2 Hz, 2H), 6.05 (s, 2H), 1.89 (s, 6H). 13C NMR (350 K, $C_2D_2Cl_4$): δ 145.0, 141.4, 141.2, 132.5, 128.8, 127.9, 126.5, 124.1, 115.0, 111.4, 15.3.

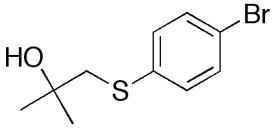
(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*)-5-(4-(methylthio)phenyl)penta-2,4-dienylidene)hex-3-enedinitrile (A2): Yield: 20%, as a purple crystalline solid. 1H NMR (CD_2Cl_2): δ 7.43 (d, J = 8.4 Hz, 4H), 7.29 (d, J = 8.1 Hz, 4H), 6.84 (m, 10H), 6.69 (s, 2H), 2.54 (s, 6H). 13C NMR (350 K, $C_2D_2Cl_4$): δ 144.5, 142.1, 139.9, 137.8, 133.3, 128.9, 128.5, 127.3, 126.7, 115.0, 111.2.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*)-7-(4-(methylthio)phenyl)hepta-2,4,6-trienylidene)hex-3-enedinitrile (A3): Yield: 16%, as a dark purple solid. 1H NMR ($C_2D_2Cl_4$): δ 8.07 (d, J = 8.6 Hz, 4H), 7.95 (d, J = 8.5 Hz, 4H), 7.66-7.21 (m, 16H), 2.72 (s, 6H). 13C NMR could not be obtained due to compound insolubility.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*,8*E*)-9-(4-(methylthio)phenyl)nona-2,4,6,8-tetraenylidene)hex-3-enedinitrile (A4): Yield: 10%, as a dark black solid. 1H NMR (500 MHz, CD_2Cl_2): δ 7.37 (d, J = 8.0 Hz, 4H), 7.22 (d, J = 7.5 Hz, 4H), 6.92-6.45 (m, 16H), 6.49 (m, 4H), 2.51 (s, 6H). 13C NMR could not be obtained due to compound insolubility.

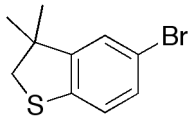


Scheme S1. Synthesis of 3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophene-5-carbaldehyde from 4-bromothiophenol.

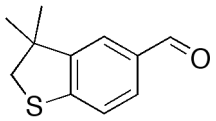


4-bromophenylthio 2-methyl-propan-2-ol: 4-bromothiophenol (1.0 g, 5.3 mmol) and 1,2-Epoxy-2-methylpropane (0.76 g, 10.5 mmol) were added to a 100-mL RBF with magnetic stir

bar. The reaction flask was then charged with 50 mL of acetone (from drum) and nitrogen was bubbled through for 20 min. Triethylamine (3 mL) was added to reaction via syringe. The reaction was stirred overnight at room temperature. The organic solvent was removed by rotary evaporation. The product was isolated by column chromatography (20% ethyl acetate in hexanes) and 1.28 g (93%) of clear oil was isolated. 1H NMR ($CDCl_3$): δ 7.43 (d, J = 8.4, 2H), 7.31 (d, J = 6.9 Hz, 2H), 3.11 (s, 2H), 2.13 (bs, 1H), 1.32 (s, 6H). 13C NMR ($CDC1_3$): δ 136.8, 132.4, 131.3, 120.4, 71.2, 48.9, 29.1.

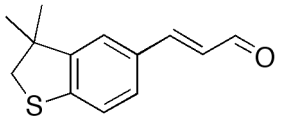


5-bromo-3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophene: A solution of 4-bromophenylthio 2-methyl-propan-2-ol (0.36 g, 1.39 mmol) CH_2Cl_2 was added to a stirring suspension of aluminum chloride (0.20 g, 1.53 mmol) in CH_2Cl_2 cool in an ice bath. The suspension was left to stir overnight while warming to room temperature. Water was added to quench unreacted $AlCl_3$. The organic layer was washed with water and then sat. aqueous sodium chloride solution before drying over $MgSO_4$. The product was isolated by column chromatography (100% hexanes) and 1.90 g (57%) of clear oil was isolated. 1H NMR ($CDCl_3$): δ 7.23 (dd, J = 1.8 Hz, 8.4 Hz, 1H), 7.14 (d, J = 1.8 Hz, 1H), 7.05 (d, J = 8.1 Hz, 1H), 3.18 (s, 2H), 1.36 (s, 6H). 13C NMR ($CDC1_3$): δ 150.7, 140.1, 130.6, 126.4, 124.1, 118.2, 47.9, 47.8, 27.6.

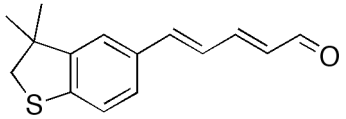


3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophene-5-carbaldehyde: A 100-mL RBF was flame dried and charged with 5-bromo-3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophene (0.310 g, 1.20 mmol) and anhydrous THF (25 mL). The reaction flask was cooled to -78° C in an acetone/dry ice bath for 40 min. 0.9 mL of n-BuLi (1.6 M in hexanes) was dripped into the reaction slowly over 5 minutes. The reaction was allowed to stir at -78° C for 30 min before dry DMF (1 mL) was added dropwise. This reaction was allowed to warm to room temperature overnight. The reaction was then quenched with water, left to stir for 10 min and extracted with ether. The organic layers were combined and washed with sat. sodium chloride solution and then dried over

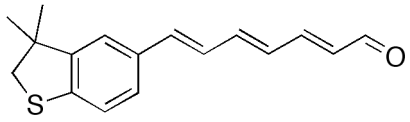
$MgSO_4$. Product was isolated by column chromatography to yield 0.203 g (94 %) of a colorless oil. NOTE: yields varied. 1H NMR ($CDCl_3$): δ 9.88 (s, 1H), 7.61 (dd, J = Hz, Hz, 1H), 7.55 (d, 1H), 7.29 (d, J = 8.1 Hz, 1H), 3.24 (s, 2H), 1.41 (s, 6H). 13C NMR ($CDC1_3$): δ 191.6, 150.5, 149.6, 134.1, 131.1, 123.0, 122.9, 47.8, 47.3, 27.9.



(*E*)-3-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)acrylaldehyde: The product, a pale yellow solid, was isolated by column chromatography (10% ethyl acetate in hexanes). Yield: 94%. 1H NMR ($CDCl_3$): δ 9.68 (d, J = 8.0 Hz, 1H), 7.44 (d, J = 15.8 Hz, 1H), 7.32 (dd, J = 1.4 Hz, 8.0 Hz, 1H), 7.22 (m, 2H), 6.68 (dd, J = 7.8 Hz, 15.8 Hz, 2H), 3.21 (s, 2H), 1.39 (s, 6H). 13C NMR ($CDC1_3$): δ 193.6, 152.8, 149.1, 145.9, 130.7, 128.3, 127.0, 122.8, 122.4, 47.5, 47.1, 27.5.

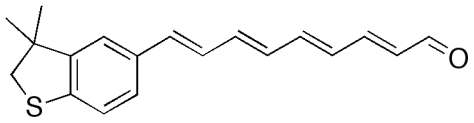


(2*E*,4*E*)-5-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)penta-2,4-dienal: Purification by column chromatography (10% ethyl acetate in hexanes). Yield: 92%, as a yellow solid. 1H NMR (400 MHz, $CDCl_3$): δ 9.61 (d, J = 8.0 Hz, 1H), 7.19 (m, 5H), 6.96 (m, 2H), 6.26 (dd, J = 8.0 Hz, 15.2 Hz, 1H), 3.22 (s, 2H), 1.40 (s, 6H). 13C NMR ($CDC1_3$): δ 193.8, 152.8, 149.3, 143.9, 142.9, 132.7, 131.2, 127.8, 125.1, 123.1, 121.8, 47.8, 47.5, 27.8.

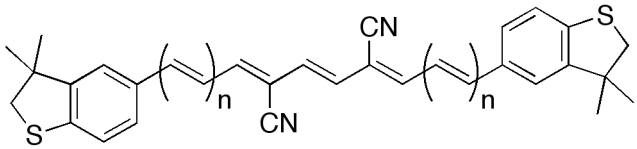


(2*E*,4*E*,6*E*)-7-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)hepta-2,4,6-trienal:
Purification by column chromatography (10% ethyl acetate in hexanes). Yield: 90%, as an orange solid. 1H NMR ($CDCl_3$): δ 9.87 (d, J = 7.8 Hz, 1H), 7.16 (m, 4H), 6.80 (m, 3H), 6.50 (dt, J = 2.7 Hz, 11.1 Hz, 1H), 6.12 (dd, J = 8.1 Hz, 15.0 Hz, 1H), 3.16 (s, 2H), 1.35 (s, 6H). 13C

NMR ($CDC1_3$): δ 193.8, 152.3, 149.4, 142.7, 138.8, 133.5, 131.2, 129.9, 127.1, 126.7, 123.0, 121.4, 47.8, 47.5, 27.8.



(2*E*,4*E*,6*E*,8*E*)-9-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)nona-2,4,6,8-tetraenal: Purification by column chromatography (10% ethyl acetate in hexanes). Yield: 63%, as a red-orange solid. 1H NMR (500 MHz, $CDCl_3$): δ 9.55 (d, J = 8.1 Hz, 1H), 7.18-7.07 (m, 4H), 6.85-6.56 (m, 4H), 6.48 (d, J = 11.1 Hz, 1H), 6.41 (dd, J = 8.1 Hz, 11.7 Hz, 1H), 6.14 (dd, J = 8.1 Hz, 15.3 Hz, 1H), 3.16 (s, 2H), 1.36 (s, 6H). 13C NMR ($CDC1_3$): δ 193.9, 152.4, 149.1, 143.3, 142.0, 139.7, 136.5, 133.9, 131.6, 131.2, 130.0, 127.4, 126.9, 123.0, 121.1, 47.8, 47.5, 27.8.



Bn; n = 0, 1, 2, 3, 4

(2*Z*,3*E*,5*Z*)-2,5-bis((3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)methylene)hex-3-enedinitrile (B0): Yield: 33%, as a lemon-yellow solid. 1H NMR ($CDCl_3$): δ 7.61 (d, J = 1.7 Hz, 2H), 7.57 (dd, J = 1.7 Hz, 8.2 Hz, 2H), 7.25 (d, J = 8.3 Hz, 2H), 7.12 (s, 2H), 6.79 (s, 2H), 3.24 (s, 2H), 1.42 (s, 12H).

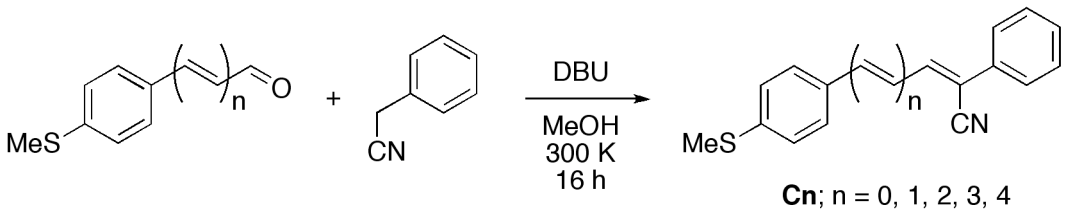
(2*Z*,3*E*,5*Z*)-2,5-bis((*E*)-3-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)allylidene)hex-3-enedinitrile (B1): Yield: 30%, as a red solid. 1H NMR (CD_2Cl_2): δ 7.33 (dd, J = 1.5 Hz, 8.1 Hz, 2H), 7.28-7.19 (m, 6H), 7.05 (d, J = 11.4 Hz, 2H), 6.98 (d, J = 15.3 Hz, 2H), 6.70 (s, 2H), 3.24 (s, 4H), 1.41 (s, 12H). 13C NMR ($CDC1_3$): δ 149.7, 146.0, 144.2, 142.2, 132.9, 129.3, 127.9, 123.8, 123.1, 122.0, 115.8, 111.3, 47.5, 47.0, 26.6.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*)-5-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)penta-2,4-dienylidene)hex-3-enedinitrile (B2): Yield: 17%, as a purple solid. 1H NMR (CD_2Cl_2): δ 7.12

(m, 6H), 7.05-6.77 (m, 10H), 6.67 (s, 2H), 3.23 (s, 4H), 1.41 (s, 12H). 13C NMR ($CDC1_3$): δ 149.2, 144.2, 142.9, 138.9, 133.6, 129.3, 128.7, 127.4, 127.1, 123.1, 121.3, 115.8, 111.3, 47.9, 47.5, 27.8.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*)-7-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)hepta-2,4,6-trienylidene)hex-3-enedinitrile (B3): Yield: 10%, as a dark solid. 1H NMR (DMSO-d_6): δ 7.44 (d, J = 11.9 Hz, 2H), 7.35 (s, 2H), 7.31 (d, J = 8.4 Hz, 2H), 7.20 (d, J = 6.6, 2H), 7.09 (dd, J = 12.3 Hz, 17.5 Hz, 2H), 6.93 (dd, J = 13.5 Hz, 16.9 Hz, 2H), 6.85-6.62 (m, 8H), 5.75 (s, 2H), 3.22 (s, 4H), 2.088 (s, 12H). 13C NMR ($CDCl_3$): δ 149.1, 145.0, 142.6, 142.1, 140.0, 136.7, 134.0, 132.1, 129.4, 128.9, 127.6, 126.9, 123.0, 121.1, 115.8, 111.3, 47.9, 47.5, 27.8.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*,8*E*)-9-(3,3-dimethyl-2,3-dihydrobenzo[*b*]thiophen-5-yl)nona-2,4,6,8-tetraenylidene)hex-3-enedinitrile (B4): Yield: 8%, as a dark solid. 1H NMR (CD_2Cl_2): δ 7.22-7.11 (m, 6H), 6.90-6.48 (m, 26H), 3.21 (s, 4H), 1.40 (s, 12H). 13C NMR (500 MHz, CD_2Cl_2): δ 148.2, 144.0, 141.6, 140.7, 138.7, 136.7, 134.4, 133.4, 131.7, 128.6, 128.0, 127.1, 125.7, 122.0, 120.2, 114.8, 110.4, 47.0, 46.5, 26.9; one carbon not observed.



Cn; n = 0, 1, 2, 3, 4

General Procedure for Single Knoevenagel Condensation with 4-methylthio-benzyl cyanide: The aldehyde (1.0 eq.) and benzyl cyanide (1.1 eq.) were placed in a 50-mL RBF with a magnetic stir bar. And were dissolved in methanol or methanol/tetrahydrofuran mixture depending on solubility of aldehehyde. The flask was sealed with a rubber septum and purged with nitrogen for 10 minutes before DBU (1.1 eq) was added via syringe and the solution was left stirring overnight. The solid product was filtered off and washed with methanol.

(*Z*)-3-(4-(methylthio)phenyl)-2-phenylacrylonitrile (C0): Yield: 75%, as a white solid. 1H NMR ($CDCl_3$): δ 7.86 (d, J = 8.4 Hz, 2H), 7.68 (dd, J = 1.5 Hz, 8.0 Hz, 2H), 7.47 (m, 4H), 7.31

(d, J = 8.5 Hz, 2H), 2.56 (s, 3H). 13C NMR ($CDCl_3$): δ 143.0, 141.9, 135.0, 130.5, 130.2, 129.4, 129.4, 126.3, 126.2, 118.6, 110.7, 15.4.

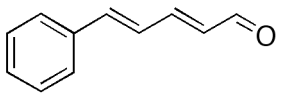
(2*Z*,4*E*)-5-(4-(methylthio)phenyl)-2-phenylpenta-2,4-dienenitrile (C1): Yield: 52%, as a yellow solid. 1H NMR ($CDCl_3$): δ 7.70 (d, J = 8.0 Hz, 2H), 7.48 (m, 7H), 7.27 (d, 2H), 7.05 (d, J = 13.7 Hz, 1H), 2.58 (s, 3H). 13C NMR ($CDCl_3$): δ 142.1, 141.4, 141.0, 133.7, 132.8, 129.5, 129.4, 128.3, 126.6, 126.0, 124.8, 117.5, 113.0, 15.7.

(2*Z*,4*E*,6*E*)-7-(4-(methylthio)phenyl)-2-phenylhepta-2,4,6-trienenitrile (C2): Yield: 43%, as an orange-yellow solid. 1H NMR ($CDCl_3$): δ 7.65 (d, J = 7.0 Hz, 2H), 7.43 (m, 6H), 7.27 (d, J = 8.5 Hz, 2H), 6.98 (m, 4H), 2.57 (s, 3H). 13C NMR (500 MHz, $CDCl_3$): δ 141.5, 141.3, 137.2, 133.4, 133.3, 129.1, 129.0, 127.4, 127.3, 126.4, 125.6, 117.1, 112.4, 101.3, 15.5.

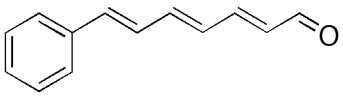
(2*Z*,4*E*,6*E*,8*E*)-9-(4-(methylthio)phenyl)-2-phenylnona-2,4,6,8-tetraenenitrile (C3): Yield: 24%, as an orange solid. 1H NMR (500 MHz, $CDCl_3$): δ 7.63 (d, J = 12.5 Hz, 2H), 7.43 (t, J = 7.0 Hz, 2H), 7.39-7.32 (m, 4H), 7.42 (d, J = 8.0 Hz, 2H), 6.91 (m, 2H), 6.78 (dd, J = 12 Hz, 14.5 Hz, 1H), 6.65 (m, 2H), 6.55 (dd, J = 11 Hz, 14.5 Hz, 1H), 2.52 (s, 3H). 13C NMR (500 MHz, $CDCl_3$): δ 141.4, 141.3, 139.0, 138.5, 135.1, 133.7, 133.4, 132.0, 129.1, 129.0, 127.9, 127.1, 126.5, 125.5, 117.1, 112.2, 15.6.

(2*Z*,4*E*,6*E*,8*E*,10*E*)-11-(4-(methylthio)phenyl)-2-phenylundeca-2,4,6,8,10-pentaenenitrile (C4): Yield: 20%, as a dark red solid. 1H NMR ($CDCl_3$): δ 7.62 (d, J = 7.1 Hz, 2H), 7.38 (m, 6H), 7.21 (d, J = 8.4 Hz, 2H), 6.85 (m, 3H), 6.66 (m, 5H), 2.52 (s, 3H). 13C NMR (500 MHz, $CDCl_3$): δ 141.4, 141.2, 138.6, 138.4, 136.5, 134.0, 133.4, 132.7, 129.04, 129.00, 128.9, 128.2, 127.0, 126.5, 125.5, 117.1, 112.1, 15.7.

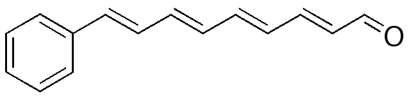
1-phenyl-enals (R = C_6H_5):



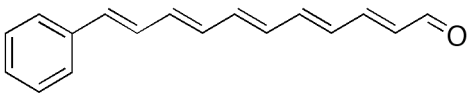
(2*E*,4*E*)-5-phenylpenta-2,4-dienal (Cont-3): Yield: 92%. 1H NMR ($CDCl_3$): δ 9.63 (d, J = 8.1 Hz, 1H), 7.51 (d, J = 8.4 Hz, 2H), 7.34 (m, 4H), 7.01 (d, J = 4.2, 2H), 6.30 (dd, J = 6.0 Hz, 11.4 Hz. 13C NMR ($CDCl_3$): δ 194.1, 152.6, 142.9, 136.0, 132.0, 130.1, 129.4, 128.0, 126.6.



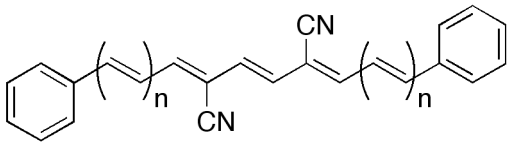
(2*E*,4*E*,6*E*)-7-phenylhepta-2,4,6-trienal: Yield: 88%, as a dark yellow solid. 1H NMR (400 MHz, $CDCl_3$): δ 9.62 (d, J = 8.0 Hz, 1H), 7.49 (d, J = 6.8 Hz, 2H), 7.40-7.28 (m, 3H), 7.23 (dd, J = 11.6 Hz, 15.2 Hz, 1H), 6.96 (m, 3H), 6.61 (dd, J = 11.2 Hz, 14.0 Hz, 1H), 6.22 (dd, J = 7.6 Hz, 15.2 Hz, 1H). 13C NMR (400 MHz, $CDCl_3$): δ 193.6, 151.8, 142.8, 138.4, 136.3, 131.2, 130.2, 128.9, 128.8, 127.7, 127.1. One carbon peak not observed.



(2*E*,4*E*,6*E*,8*E*)-9-phenylnona-2,4,6,8-tetraenal: Yield: 70%, as an orange solid. 1H NMR ($CDCl_3$): δ 9.59 (d, J = 8.1 Hz, 1H), 7.45 (d, J = 7.2 Hz, 2H), 7.37-7.24 (m, 2H), 7.17 (dd, J = 11.4 Hz, 15.0 Hz, 1H), 6.90-6.42 (m, 6H), 6.19 (dd, J = 8.1 Hz, 15.0 Hz, 1H). 13C NMR ($CDCl_3$): δ 194.0, 152.2, 143.0, 139.3, 137.1, 136.5, 132.3, 131.4, 130.4, 129.2, 128.8, 127.2. One carbon not observed.



(2*E*,4*E*,6*E*,8*E*,10*E*)-11-phenylundeca-2,4,6,8,10-pentaenal: Yield: 55%, as a dark orange solid. 1H NMR ($CDCl_3$): δ 9.61 (d, J = 7.2 Hz, 1H), 7.46 (d, J = 7.5 Hz, 2H), 7.35 (t, J = 8.1 Hz, 1H), 7.27-7.13 (m, 2H), 6.94-6.37 (m, 8H), 6.20 (dd, J = 8.1 Hz, 15.0 Hz, 1H). 13C NMR ($CDCl_3$): δ 196.4, 154.7, 145.6, 141.8, 140.0, 139.7, 135.5, 134.7, 133.8, 132.8, 131.9, 131.6, 131.0, 129.5. One carbon not observed.



Dn; n = 0, 1, 2, 3, 4, 5

(2*Z*,3*E*,5*Z*)-2,5-dibenzylidenehex-3-enedinitrile (D0): Yield: 55%, as a yellow crystalline solid. 1H NMR ($C_2D_2Cl_4$): δ 7.87-7.84 (m, 4H), 7.47 (dd, J = 1.8 Hz, 5.0 Hz, 6H), 7.22 (s, 2H), 6.87 (s, 2H). 13C NMR (350 K, $C_2D_2Cl_4$): δ 145.6, 13.1, 131.1, 130.1, 129.3, 129.0, 116.0, 109.3.

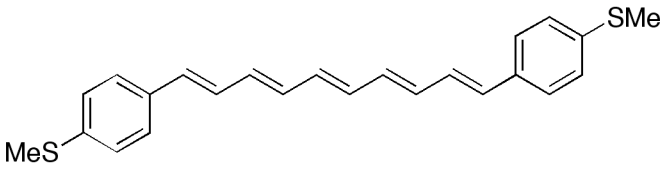
(2*Z*,3*E*,5*Z*)-2,5-bis((*E*)-3-phenylallylidene)hex-3-enedinitrile (D1): Yield: 60%, as an orange crystalline solid. 1H NMR ($C_2D_2Cl_4$): δ 7.59 (d, J = 6.6 Hz, 4H), 7.44-7.28 (m, 8H), 7.08-7.01 (m, 4H), 6.76 (s, 2H). 13C NMR (350 K, $C_2D_2Cl_4$): δ 145.0, 142.0, 135.6, 129.7, 129.0, 128.8, 127.5, 124.7, 114.8, 111.9.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*)-5-phenylpenta-2,4-dienylidene)hex-3-enedinitrile (D2): Yield: 30%, as a red solid. 1H NMR (400 MHz, 350 K, DMSO-d_6): δ 7.59 (d, J = 7.6 Hz, 4H), 7.44-7.30 (m, 8H), 7.24 (dd, J = 10.8 Hz, 15.6 Hz, 2H), 7.04-6.95 (m, 4H), 6.84 (dd, J = 11.6 Hz, 14.0 Hz, 2H), 6.75 (s, 2H). 13C NMR (370 K, DMSO-d_6): δ 146.2, 143.4, 138.8, 137.1, 129.22, 129.20, 129.15, 129.12, 127.6, 115.5, 111.0.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*)-7-phenylhepta-2,4,6-trienylidene)hex-3-enedinitrile (D3): Yield: 30%, as a dark purple solid. 1H NMR (400 MHz, 350 K, DMSO-d_6): δ 7.53 (d, J = 7.6 Hz, 4H), 7.39-7.27 (m, 8H), 7.10-6.68 (m, 14H). 13C NMR (380 K, DMSO-d_6): δ 146.2, 143.5, 140.0, 137.8, 136.6, 133.4, 129.8, 129.5, 129.3, 128.9, 127.6, 115.9, 111.3.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*,8*E*)-9-phenylnona-2,4,6,8-tetraenylidene)hex-3-enedinitrile (D4): Yield: 22%, as a dark purple-black solid. 1H NMR (400 MHz, 350 K, $C_2H_2Cl_4$): δ 7.47 (d, J = 7.6 Hz, 4H), 7.37 (t, J = 7.2 Hz, 4H), 7.28 (t, J = 7.2 Hz, 2H), 6.95-6.47 (m, 18H). 13C NMR (400 MHz, 350 K, $C_2D_2Cl_4$): δ 144.0, 141.6, 136.6, 136.4, 134.4, 132.3, 131.7, 128.6, 128.4, 128.2, 127.5, 126.1. Due to insolubility four carbons were not observed in 13C NMR.

(2*Z*,3*E*,5*Z*)-2,5-bis((2*E*,4*E*,6*E*,8*E*,10*E*)-11-phenylundeca-2,4,6,8,10-pentaenylidene)hex-3-enedinitrile (D5): Yield: 10%, as a black solid. 1H and 13H NMR data unavailable due to insolubility. See crystal structure determination section of supporting information.



(1*E*,3*E*,5*E*,7*E*,9*E*)-1,10-bis(4-(methylthio)phenyl)deca-1,3,5,7,9-pentaene (Cont-1): Compound was made according to previously reported procedure by Spangler *et al.*[1]

II. Crystal Structure Determination.

Crystals of **D0-D1** and **D2-D5** suitable for single-crystal XRD were prepared by bi-layer diffusion of CH_3OH into a solution of CH_2Cl_2 and evaporation from a 1:1 solution of CH_2Cl_2/hexanes, respectively. The former yielded cubic crystals while the latter co-crystallized with one solvent molecule (CH_2Cl_2) into thin, dark-colored needles.[3]

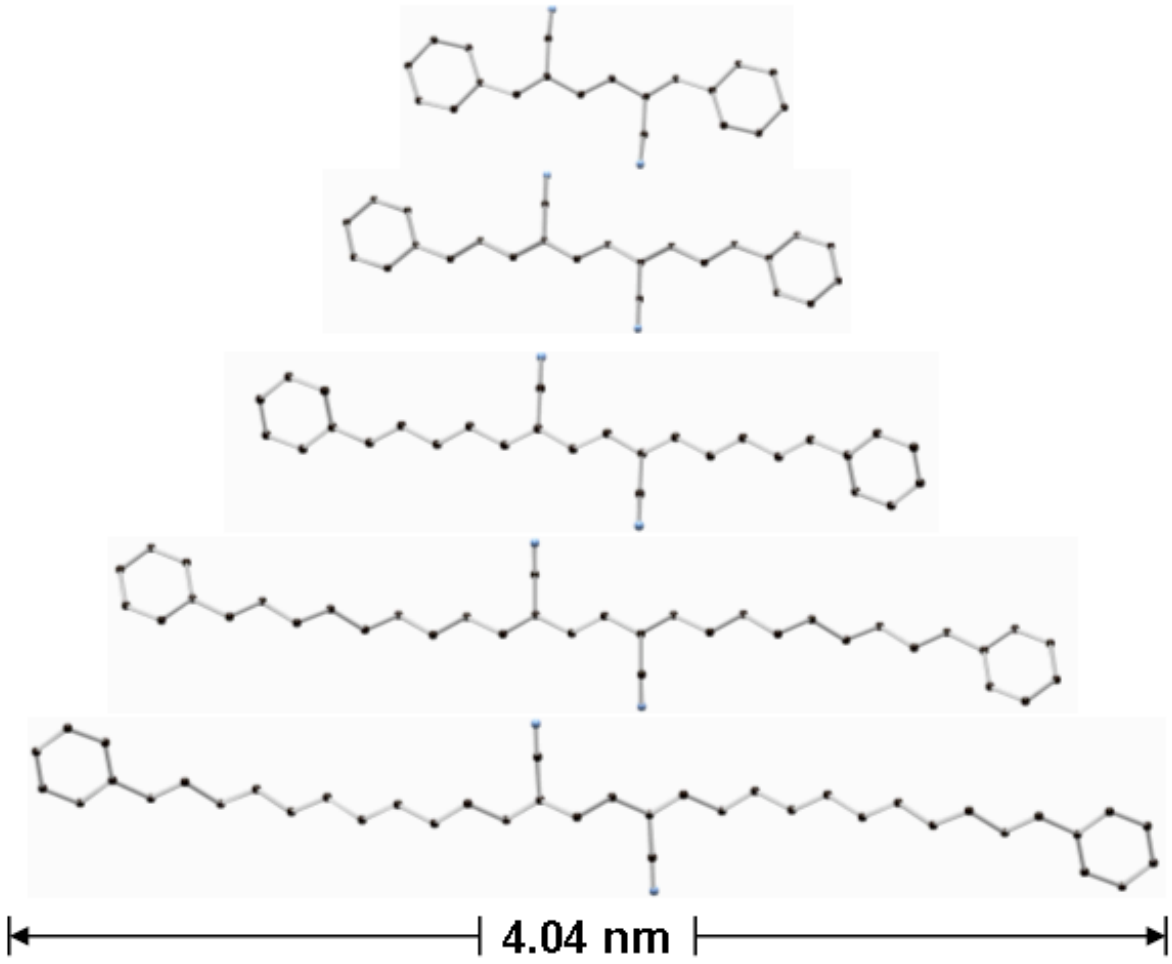


Figure S1. Structure determined for the **Dn** series from X-ray crystallography.

III. UV/vis Absorption Spectrometry:

General Procedure and Instrumentation: Absorption spectra were taken on an Agilent Technologies 8453 UV/vis spectrophotometer. Solutions of oligoenes were prepared with dichloromethane as solvent.

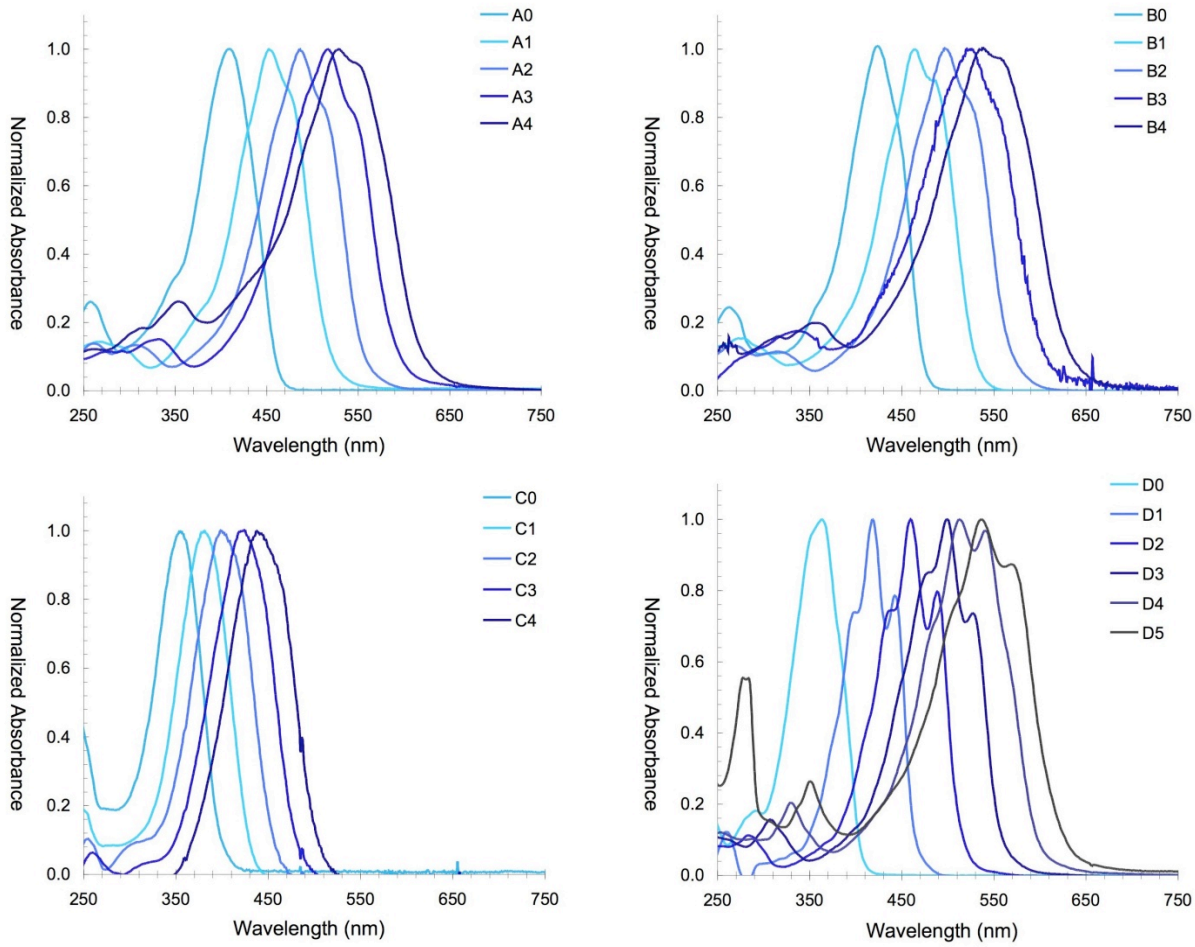


Figure S2. Optical absorption spectra of series (A) **An**, (B) **Bn**, (C) **Cn**, and (D) **Dn** in dichloromethane. Strongest wavelength absorptions have been normalized to an absorbance of 1.0.

Figure S3. Photograph of dilute solutions (~10^{-3} M) of oligoene series **An** (top) and **Dn** (bottom) in dichloromethane. The HOMO-LUMO gap energy is a function of molecular length.

III. Cyclic Voltammetry:

General Procedure and Data Analysis: Cyclic voltammertry (CV) was preformed using a CHI600c potentiometer. All electrodes were purchased from Bioanalytical Systems, Inc: working electrode was a glass carbon electrode (MF-2070), reference electrode was Ag/AgCl (MF-2062), and auxiliary electrode was a platinum electrode (MW-4130). All measurements were carried out under argon in dimethylformamide solution with tetrabutylammonium hexafluorophosphate electrolyte (0.1 M) and oligoene (350 μM). Ferrocene was used as an internal standard.

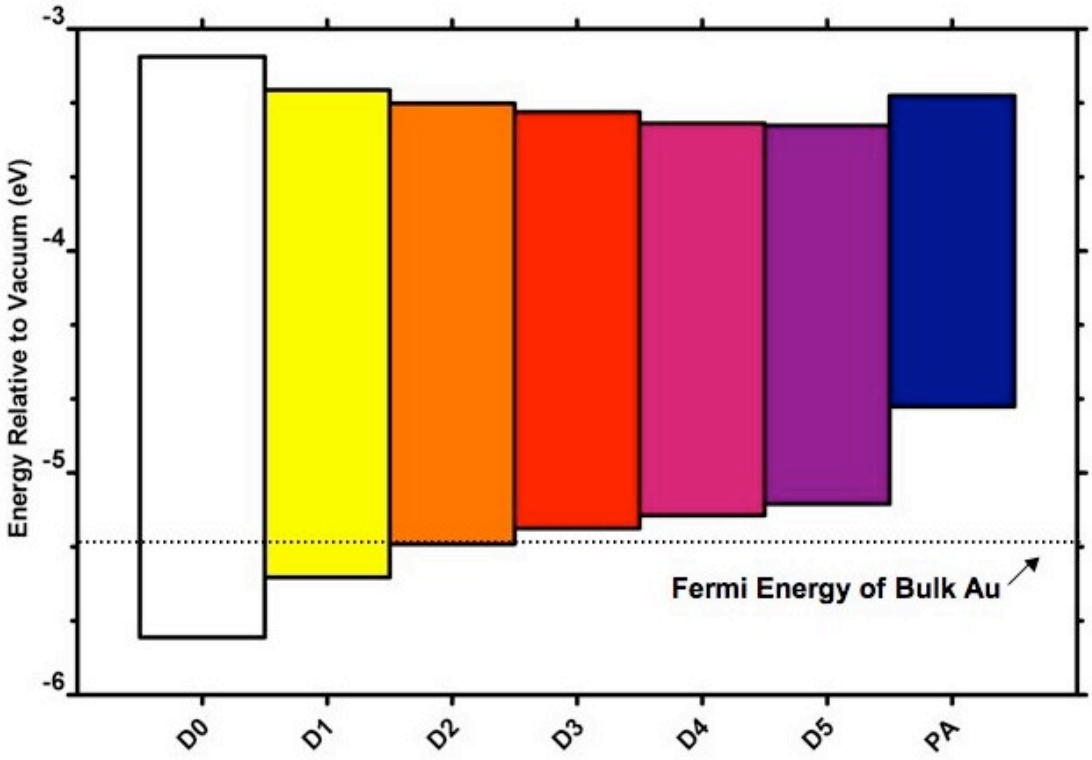


Figure S4. Ionization potentials, obtained from CV, outline the HOMO/LUMO gap energies relative to vacuum for nitrile-functionalized oligoenes (**Dn**). Nitriles functionalization lowers the energy of both the LUMO (up to 0.68 eV) and the HOMO (up to 0.44 eV) compared to unfunctionalized diphenyl-oligoenes. Reported values for the PA (polyacetylene) and the Fermi level of bulk Au are included for comparison.

IV. Modified STM Break Junction Studies:

Experimental Procedure and Data Analysis:

The details of our experimental setup have been described previously.[3] Briefly, we prepare gold samples by evaporating 100 nm of gold onto freshly cleaved mica. During measurement, the sample is mounted on top of a single-axis piezoelectric positioner below a hand-cut gold tip in a home-built STM setup. The sample-tip junction is stretched and compressed with sub-nanometer precision by moving the substrate relative to the tip at a rate of 15 nm/s with the piezoelectric (Mad City Labs) while applying constant bias to the sample through a series resistor. The current in the tip is captured by a Keithly 428 current-voltage amplifier. The sample position is manipulated and data acquired at 40 kHz using a data acquisition board (National Instruments, PXI-4461) and custom-built software written in Igor (Wavemetrics, Inc). All position determinations were based on measurements with a built-in position sensor within our custom piezoelectric positioner. This position sensor was calibrated both by the manufacturer and by us using laser interference measurements. We found the absolute values of the measured displacements to be accurate to within 5%.

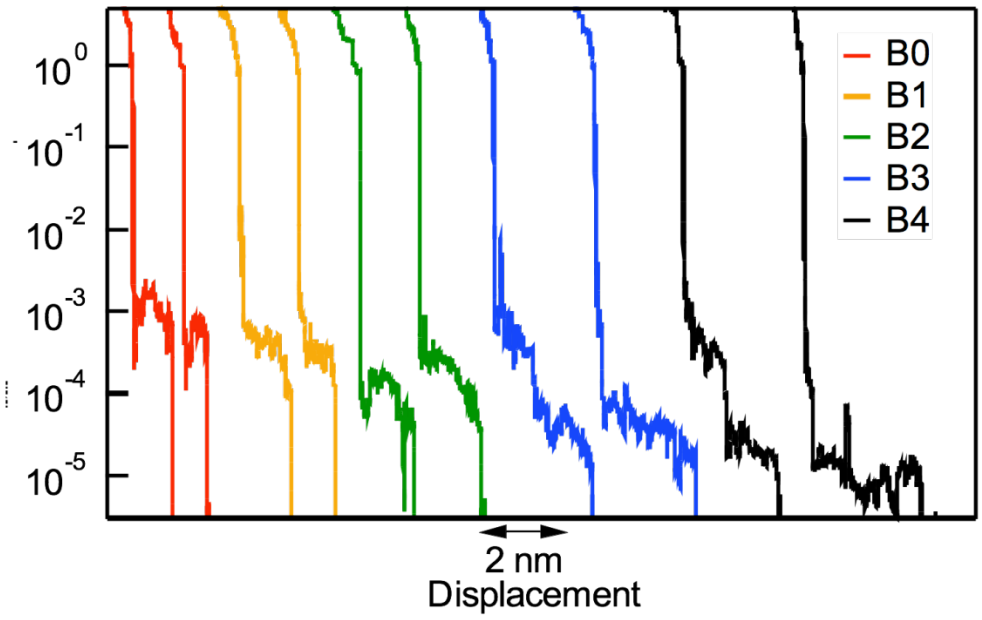


Figure S5. Sample conductance traces gained from STM-based break junction measurements on **Bn** series. Step lengths become longer as molecular length increases, showing that conductive junctions are sustained at larger electrode separations with longer molecules.

We form metal-molecule-metal junctions by smashing the tip and substrate together until conductance exceeds 5 G_0 and then pulling them apart. All conductance traces acquired that reach a conductance below 5e-6 G_0 are then added to a linear binned histogram by an automated algorithm without any further data selection. Typically, 5000 traces are used to construct conductance histograms.

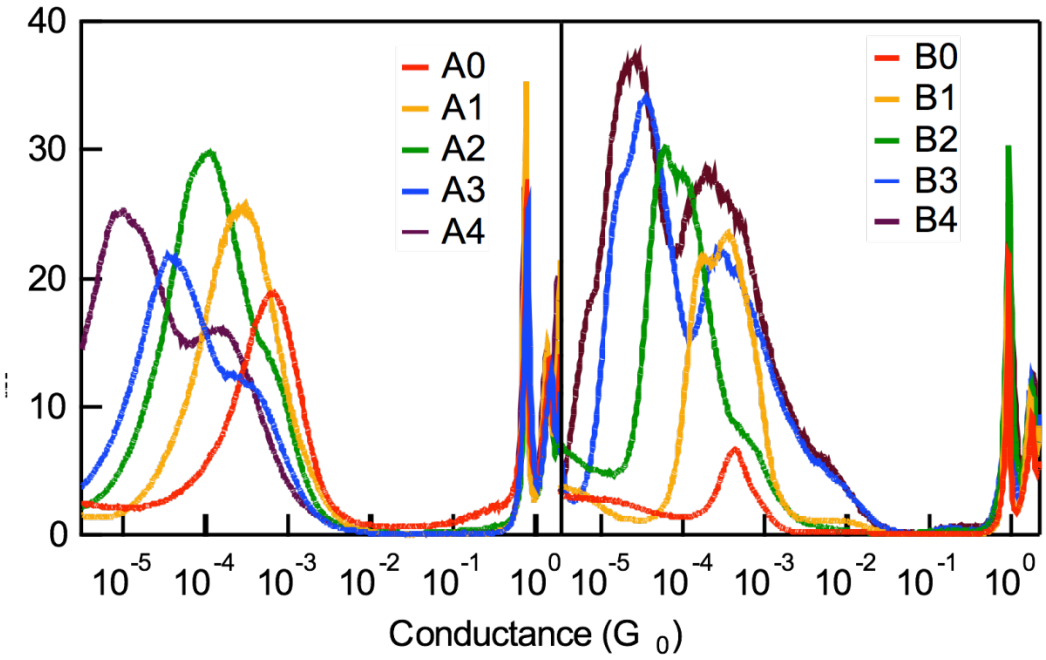


Figure S6. Logarithm binned conductance histograms for **An** and **Bn** series. Bin size is 100 per decade.

Two-dimensional histograms are automatically generated[4,5] with the added requirement that a G_0 break is clearly identifiable in the trace (more than 90% of traces that start with a conductance greater than 1 G_0 and break satisfy this requirement). In two-dimensional histograms conductance is binned logarithmically with 100 bins per decade, while displacement is binned linearly for image clarity.

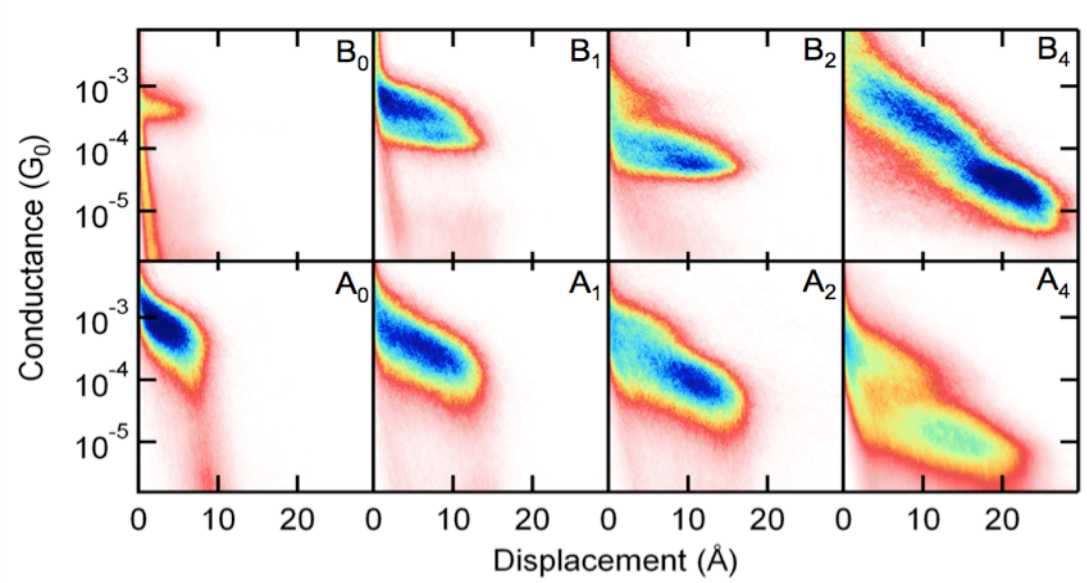


Figure S7. Two-dimensional histograms for the **An** and **Bn** series. The displacement axis has linear bins while the conductance axis has logarithm bins (100 per decade). The higher conductance regime lengthens as the oligomer length increases.

We have not observed that cyano groups bind to gold. 1,4-dicyanobenzene, 1,4-dicyano-2-butene and oligoenes **Dn** do not show conductance peaks and likely do not form molecular junctions. Like the cyano-functionalized oligoenes, oligoenes without cyano groups (**Cont-1**) contain 2 conductance peaks demonstrating that cyano groups do not play a direct role in junction formation.

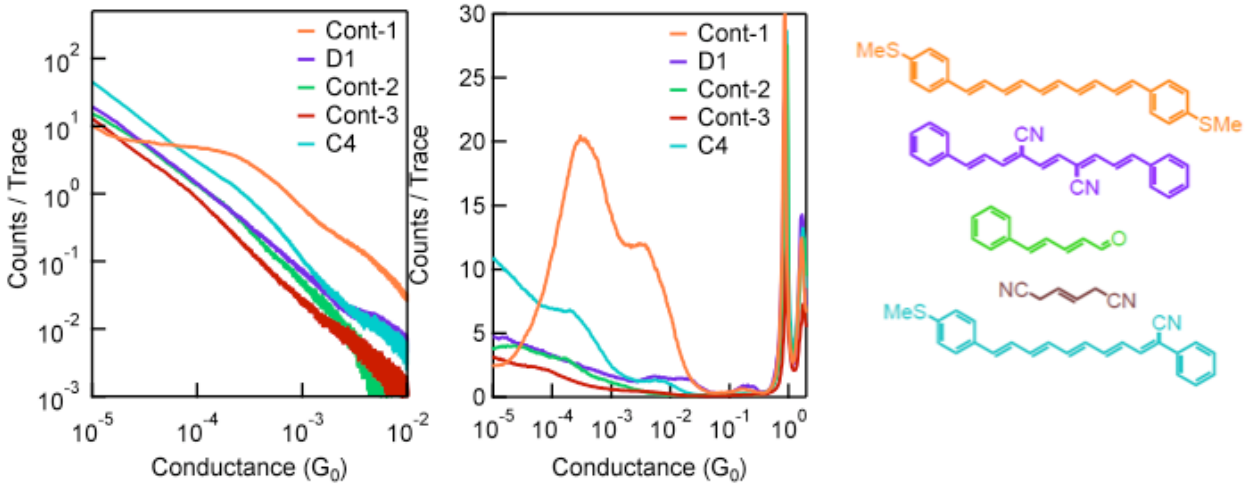


Figure S8. Linear and log binned conductance histograms of control compounds investigated. Structures are shown on the right.

Analysis of Traces with Zig-Zag Ramp: Traces were selected from all measured traces using an automated algorithm. The number of data points in the initial "hold" region of each trace (starting at time t=0s) that had a conductance within the required range was determined. Typically, 50% of all measured traces had a molecule during the initial "hold" region. All selected traces were used to construct a two-dimensional conductance vs. time histograms. Control measurements were carried out with 1,6-bis(methylsulfide)hexane, a saturated molecule. The zig-zag ramp used was chosen so that the proportional change in the in electrode-electrode separation relative to the length of the molecule was the same as in the ramp used in Figure 4 of the manuscript. (Figure S9. The probability of having a junction start at the selected conductance with hexane is 15%, which is significantly lower than the 50% success rate with **B4**. In addition, no reproducible modulation in conductance during the ramp is observed. Traces collected without the presence of any molecules showed conductance oscillating between a closed and an open junction during the zig-zag ramp.

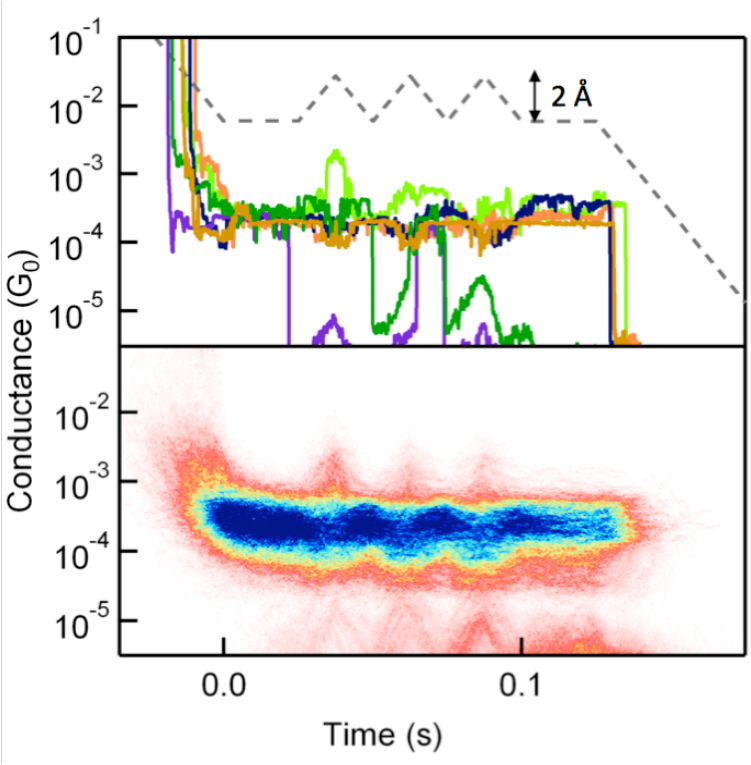


Figure S9. Upper panel: sample traces measured using the modified piezo ramp with a 2Å amplitude with 1,6-bis(methylsulfide)hexane. Lower panel: two –dimensional histograms of all selected traces (1477 out of 10000) measured with the zig-zag ramp.

V. Theoretical Methods and Details.

All electronic structure calculations used Jaguar (version 7.0, Schrodinger LLC, New York, NY, 2007) using the B3LYP hybrid functional and the 6-31G** basis sets. The Au atoms were described using the LACVP local potentials. The geometries of **A1**, **C4**, **Au_2-A1**, and **Au_2-C4** were fully optimized. The final geometries and total energies are given below.

Molecule: **A1**

DFT: B3LYP, 6-31G**

Final total energy: -1909.86419 hartrees

Final geometry:

	angstroms		
atom	x	y	z
C1	-0.0054586777	0.0160760258	-0.0169920773
C2	0.0027896249	0.0085385018	1.3914265084
C3	1.2617271719	0.0012497986	2.0283492339
C4	2.4438887757	0.0015085903	1.3053981250
C5	2.4188241032	0.0089772276	-0.0998196412
C6	1.1742433465	0.0163070722	-0.7499110173
C7	-1.2021440991	0.0072778950	2.2052531568
C8	-2.4927133813	0.0169740384	1.7749831566
C9	-3.6075929604	0.0127547254	2.6676183755
C10	-4.9334990519	0.0224029486	2.3021511411
C11	-5.2751397474	0.0392694232	0.9081509595
N12	-5.5621980629	0.0531364751	-0.2201717100
C13	-6.0101421145	0.0154862932	3.2699705747
C14	-7.3322358085	0.0233110312	2.9653261724
C15	-8.4151733561	0.0145807935	3.9266602800
C16	-8.0813278759	-0.0032513273	5.3218234957
N17	-7.7896758628	-0.0176148026	6.4488742453
C18	-9.7352003865	0.0222806138	3.5418266357
C19	-10.8782093540	0.0136783650	4.3994222849
C20	-12.1461860159	0.0220303112	3.9044895871
C21	-13.4021959869	0.0146818583	4.6351965919
C22	-14.6079289886	0.0263155388	3.9099862826
C23	-15.8483278911	0.0202660156	4.5387136047
C24	-15.9277001203	0.0017264220	5.9370020083
C25	-14.7299066618	-0.0103240334	6.6793230835
C26	-13.4999714035	-0.0041483078	6.0445455925
H27	-3.3938194979	0.0003963107	3.7351203893
H28	-9.9256264741	0.0358768554	2.4698511737
H29	-5.7046635080	0.0023889406	4.3139517264
H30	-7.6345826850	0.0364930545	1.9204393953
H31	-1.0316634905	-0.0025146717	3.2818925770
S32	3.9910988563	0.0084039910	-0.9274374134
H33	3.3957627930	-0.0044316048	1.8287255893
H34	1.3047841869	-0.0047694344	3.1145542437
H35	-0.9481970700	0.0214885691	-0.5550998814
H36	1.1158745968	0.0221111506	-1.8319398186
H37	-10.7015210857	-0.0001287768	5.4719983301

H38	-12.2526442300	0.0358262463	2.8196296730
S39	-17.4430232213	-0.0078399861	6.8645500296
H40	-16.7451674257	0.0300240106	3.9305929385
H41	-14.5699844983	0.0404738872	2.8235381871
H42	-12.6003534518	-0.0141466040	6.6514829712
H43	-14.7718464591	-0.0246611109	7.7647246498
H44	-2.7134702204	0.0283108250	0.7108404068
C45	-18.7342463992	0.0135341018	5.5795904309
H46	-19.6832765180	0.0068539840	6.1193210506
H47	-18.6792182189	0.9193772498	4.9717731839
H48	-18.6821711952	-0.8737282692	4.9446992281
C49	3.5500383513	0.0142213788	-2.6948981937
H50	4.5004270922	0.0121132100	-3.2322812278
H51	2.9861342170	-0.8798300083	-2.9698160773
H52	2.9926291711	0.9135017312	-2.9658734422

Molecule: **C4**

B3LYP, 6-31G**

Final total energy: -1380.11453 hartrees

Final geometry:

	angstroms		
atom	x	y	z
C1	-0.0221616271	0.0305191350	0.0657841755
C2	0.0505512825	0.0074467812	1.4712469394
C3	1.3277432101	-0.0214717221	2.0644324619
C4	2.4824111277	-0.0279251694	1.2886342811
C5	2.3940480177	-0.0052338919	-0.1022654048
C6	1.1349232644	0.0236065508	-0.7067546243
C7	-1.1500491919	0.0210224430	2.3322139375
C8	-2.4642172798	0.0200794862	1.9282325595
C9	-3.5626572514	0.0607782631	2.8408985884
C10	-4.8923875129	0.0687414444	2.5311564879
C11	-5.8881283931	0.1410966200	3.5564740442
C12	-7.2395506329	0.1665423464	3.3755156381
C13	-8.1765498464	0.2718719279	4.4542673537
C14	-9.5285250050	0.3102243306	4.2922088751
C15	-10.4967098987	0.4474326670	5.3441060724
C16	-11.8299166562	0.4874863103	5.0879124803
C17	-12.9519267843	0.6487222766	6.0013380875
C18	-12.8435524243	0.8223418732	7.3944075692
C19	-13.9702420162	0.9877303016	8.1954721673
C20	-15.2552778560	0.9855665195	7.6300934672
C21	-15.3785559738	0.8073841152	6.2428590091
C22	-14.2523666593	0.6442161835	5.4547805575
H23	-7.7653337333	0.3339907054	5.4618309675
H24	-5.2159161072	0.0299975693	1.4910745619
H25	-9.9250838102	0.2437572817	3.2783197951
H26	-3.3058197567	0.0970533638	3.8970299195
H27	-7.6416884426	0.1163793456	2.3633494258
H28	-5.5136371922	0.1890132931	4.5791158155
H29	-12.1223322999	0.3951920445	4.0415861458
S30	-16.7774130997	1.1948086860	8.5286893528
H31	-16.3632711996	0.8015210691	5.7840625660
H32	-14.3719619379	0.5107655764	4.3826476703
H33	-11.8645342439	0.8327906750	7.8648242297
H34	-13.8376736657	1.1201628395	9.2633471263
H35	-2.6853256881	-0.0050533406	0.8644066396
C36	-0.9120067742	0.0384788820	3.7478490561
H37	3.2935780054	-0.0090819695	-0.7105257126

H38	3.4529261387	-0.0501423745	1.7755083334
H39	1.4134213558	-0.0385732287	3.1459155885
H40	-0.9857224925	0.0579577247	-0.4330746723
H41	1.0535213299	0.0430039393	-1.7896751657
H42	-10.1230897813	0.5277065949	6.3638829573
N43	-0.7363902581	0.0501750068	4.8991437366
C44	-16.2411684638	1.4463029825	10.2504037037
H45	-17.1591222690	1.5911879998	10.8236836060
H46	-15.7157666983	0.5714214952	10.6400607049
H47	-15.6177881406	2.3377946450	10.3492541465

Molecule: **Au_2-A1**

B3LYP, 6-31G**/LACVP

Final total energy: -2180.81428 hartrees

Final geometry:

	angstroms		
atom	x	y	z
C1	-0.1516373949	0.1679875211	-0.0920807598
C2	-0.1554448482	0.1426712864	1.3189390134
C3	1.0950682395	0.0959268006	1.9662913092
C4	2.2834179216	0.0722650893	1.2466190464
C5	2.2685631129	0.0939093271	-0.1578716415
C6	1.0283367266	0.1450026652	-0.8179739000
C7	-1.4298578120	0.1556168426	2.0175984357
C8	-1.6479765191	0.1579270279	3.3611422960
C9	-2.9622376371	0.1481850783	3.9206759664
C10	-3.2784332203	0.1451744227	5.2595695653
C11	-2.2256593264	0.1715243113	6.2341693894
N12	-1.3825923233	0.1950646990	7.0369549542
S13	3.7168477442	0.0609847888	-1.1863319944
C14	5.0901887530	-0.1068559426	-0.0023258630
C15	-4.6464451705	0.1045807583	5.7315867670
C16	-5.0255135231	0.0790647660	7.0345937175
C17	-6.3937433972	0.0239428273	7.5051142778
C18	-7.4441699552	-0.0080536046	6.5279112199
N19	-8.2840057906	-0.0353541022	5.7221000288
C20	-6.7115130661	-0.0132685978	8.8430009027
C21	-8.0256896009	-0.0812680369	9.4003620940
C22	-8.2437773535	-0.1239383696	10.7434470448
C23	-9.5174855466	-0.1938231748	11.4406077574
C24	-10.7729390874	-0.2106831967	10.7900000803
C25	-11.9527239137	-0.2787178759	11.5105828581
C26	-11.9345078346	-0.3325500112	12.9174044466
C27	-10.7015192229	-0.3151262038	13.5799404261
C28	-9.5202313733	-0.2476393486	12.8466948642
S29	-13.5277210425	-0.3528084540	13.7409995325
C30	-13.0603518622	-0.3164940106	15.5108257009
Au31	-14.5024901338	-2.6095863740	13.4571867019
H32	3.2218390586	0.0336526329	1.7879740759
H33	1.1452791694	0.0736161875	3.0507683321
H34	-1.0999330274	0.2039259268	-0.6228532913
H35	0.9941922861	0.1605641450	-1.9039558313
H36	-2.3072802772	0.1596859296	1.3702118362
H37	-0.8108994067	0.1618803954	4.0550936332

H38	-3.7981752710	0.1291394194	3.2228255261
H39	-5.4087535267	0.0872652434	4.9556374629
H40	-4.2630557325	0.0899286556	7.8105133488
H41	-5.8768160363	0.0099773858	9.5422295413
H42	-8.8616179730	-0.1007964511	8.7051387280
H43	-7.3670894811	-0.1046560205	11.3914968636
H44	-8.5712925854	-0.2372100081	13.3777291444
H45	-10.6450344931	-0.3569231448	14.6612106335
H46	-12.9045312944	-0.2987525020	10.9871035551
H47	-10.8253190860	-0.1688656503	9.7062787616
H48	5.1557753051	0.7553652430	0.6654996964
H49	5.0082487114	-1.0296409468	0.5767940192
H50	5.9958057167	-0.1510484786	-0.6111780022
H51	-12.4899264306	-1.2026905582	15.7924293051
H52	-14.0044300352	-0.3082362637	16.0571946543
H53	-12.5005591224	0.5952144728	15.7349863339
Au54	-15.6041498559	-4.9343391061	13.2321392566

Molecule: **Au_2-C4**

B3LYP, 6-31G**/LACVP

Final total energy: -1651.074011 hartrees

Final geometry:

	angstroms		
atom	x	y	z
C1	-0.0157017963	0.0713846205	0.0100088942
C2	-0.0118780466	0.0574814708	1.4167733522
C3	1.2404537151	0.0046851532	2.0705008407
C4	2.4275782274	0.0119996515	1.3502056867
C5	2.3943929024	0.0397774758	-0.0495957415
C6	1.1662093791	0.0546989829	-0.7199962650
C7	-1.2925818092	0.1475882933	2.1042742997
C8	-1.5445809720	0.0547421499	3.4348586239
C9	-2.8580297143	0.2437167487	3.9826887694
C10	-3.1844232823	0.1319396223	5.3000704689
C11	-4.4990801142	0.3756221425	5.8111019268
C12	-4.8574738253	0.2164532870	7.1164697956
C13	-6.1630000042	0.4864148915	7.6337069042
C14	-6.5143059703	0.2593894971	8.9324159169
C15	-7.8043499020	0.5445691560	9.4770163200
C16	-8.2013856165	0.2527364866	10.7570880070
C17	-7.2855822889	-0.4687689976	11.5941527484
N18	-6.5455373448	-1.0662493374	12.2662795552
S19	3.8714400398	0.2111589482	-1.0640110846
C20	4.9903736624	-1.0844467160	-0.4077782114
C21	-9.5126099593	0.6012157947	11.3377247039
C22	-10.3310782986	1.5966127061	10.7696238294
C23	-11.5618579051	1.9129444833	11.3374705296
C24	-12.0029360574	1.2502985388	12.4839933743
C25	-11.1987587534	0.2660184912	13.0606093806
C26	-9.9667531961	-0.0537118197	12.4969875873
H27	-2.4147493587	-0.1571741925	6.0159303128
H28	-6.9079680580	0.8887811008	6.9464122007
H29	-3.6437981061	0.5070433353	3.2738701458
H30	-5.7686471574	-0.1722184270	9.5981027247
H31	-5.2542664826	0.7082690098	5.0980360509
H32	-4.1091763819	-0.1462937171	7.8217503301
H33	-2.1424095822	0.3253711826	1.4455936853
H34	1.1353606077	0.0733025681	-1.8036737409
H35	-0.9674620536	0.1073238813	-0.5168104552
H36	1.2889975403	0.0083579577	3.1536287791
H37	3.3765622903	0.0336033088	1.8790898024

H38	-8.5304716919	1.0212860850	8.8235923062
H39	-12.9611702550	1.5060690611	12.9275935091
H40	-11.5281163260	-0.2545725451	13.9542156847
H41	-9.3493007561	-0.8221582988	12.9532240181
H42	-9.9947464398	2.1452441082	9.8961649563
H43	-12.1750177976	2.6907268560	10.8898848717
H44	-0.7430734747	-0.1635173370	4.1387137927
H45	5.9164448599	-0.9988878072	-0.9785381181
H46	5.2009031202	-0.9300127831	0.6503336885
H47	4.5364919704	-2.0624752090	-0.5760763271
Au48	4.7641825117	2.3655582803	-0.2666378145
Au49	5.5058118799	4.6126128264	0.7780153528

VI. References and Notes

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4. Kamenetska, M.; Koentopp, M.; Whalley, A.; Park, Y. S.; Steigerwald, M.; Nuckolls, C.; Hybertsen, M.; Venkataraman, L. *Physical Review Letters,* **2009,** 102, (12), 126803.

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