Electronic Supplementary Information for

Chiral Polymer Hosts for Circularly Polarized

Electroluminescence Devices

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CONTENTS

Experimental Details		
Fig. S1	Photoluminescence decay traces for thin films of JY_n polymers spincast on quartz	
	substrates recorded at wavelengths of 380 nm (a) and 420 nm (b) after pulsed laser	
	photoexcitation of 332 nm (temporal resolution = 25 ps).	
Fig. S2	(a) Comparisons of the UV-vis absorption (top), ECD (middle), and the Kuhn	
	dissymmetry factor (g_{abs}) spectra of a 100 μM solution of ${\bf 2}$ (black curves) and a 0.085	
	mg mL $^{-1}$ solution of JY_{100} (red curves) in 1,2-dichloroethane. (b) Comparisons of the	
	UV–vis absorption (top), ECD (middle), and the Kuhn dissymmetry factor ($g_{ m abs}$) spectra	
	of JY $_{100}$ solutions obtained with increased concentrations (0.017–0.17 mg mL $^{-1}$ in 1,2-	
	dichloroethane).	
Fig. S3	ECD spectra of a 0.028 mg mL $^{-1}$ solution of JY $_{100}$ (1,2-dichloroethane) recorded upon	
	cooling (a) and heating (b).	

Fig. S4Plots of the Kuhn dissymmetry factor (g_{abs}) as a function of wavelength for dropcast filmsof N-ethylcarbazole doped with varying concentrations of 2 (0.5–15 wt %).

Fig. S5 ECD spectra of the neat films of JY_n polymers cast on quartz substrates.

- **Fig. S6** (a) Plots of the Kuhn dissymmetry factor (g_{abs}) as a function of wavelength for the neat films of the JY_n polymers doped with 10 wt % (*R*)-Pt. (b) Plot of g_{abs} (λ_{obs} = 372 nm) as a function of the mole fraction of the chiral repeat unit (i.e., $[M^*] / ([M^*] + [M])$) in JY_n.
- Fig. S7 (a) UV–vis absorption (top) and ECD (bottom) spectra of 10 μM solutions of (*R*)- and (*S*)-Pt (1,2-dichloroethane). (b) Normalized photoluminescence spectra of 10 μM solutions of (*R*)- and (*S*)-Pt (1,2-dichloroethane) recorded upon photoexcitation at 400 nm.
- Fig. S8 g_{PL} spectra of a 100 mM (R)-Pt in 1,2-dichloroethane (1,2-DCE; red symbols) and JY0(empty blue symbols) and JY100 (filled blue symbols) films doped with 30 wt % (R)-Pt.
- **Fig. S9** (a,b) Photoluminescence spectra ($\lambda_{ex} = 339$ nm) obtained for JY₀ (a) and JY₁₀₀ (b) films doped with various concentrations (0–10 wt %) of (*S*)-Pt. (c,d) Photoluminescence decay traces ($\lambda_{obs} = 420$ nm) of JY₀ (c) and JY₁₀₀ (d) films doped with various concentrations (0–10 wt %) of (*S*)-Pt obtained after picosecond pulsed laser photoexcitation at 377 nm (temporal resolution = 25 ps).
- Fig. S10Photoluminescence decay traces of 10 wt % (S)-Pt:JY0 films (a) and 10 wt % (S)-Pt:JY100films observed at wavelengths of 420 (filled symbols; i.e., host emission) and 590 nm(empty symboles; i.e., dopant emission) after picosecond pulsed laser photoexcitation at377 nm.
- Fig. S11
 Cyclic (solid curve) and differential pulse (dotted line) voltammograms of a 0.39 mg mL⁻¹

 solution of PVK (Ar-saturated THF) containing 0.10 M Bu₄NPF₆ supporting electrolyte.
- **Fig. S12** Electroluminescence spectra of the PLEDs employing (R)-Pt:JY₀ with different doping concentrations of (R)-Pt (10, 20, and 30 wt %) as emitting layers.
- **Fig. S13** g_{PL} spectra of the 30 wt %-doped films measured with the substrates facing the PEM, showing that the signs of g_{PL} are the same as those of the measurements with the sample films facing the PEM.
- Fig. S14 Schematic diagram of the home-made CPL spectroscopic system.
- **Fig. S15** g_{EL} spectra and $\cos(\Delta\theta)$ plots of the PLEDs with a (*R*)- or (*S*)-Pt:JY_n emitting layer, showing all data points used to plot Fig. 9e.
- **Fig. S16** g_{EL} spectra and $\cos(\Delta \theta)$ plots of the PLEDs with a (*R*)- or (*S*)-Pt:JY_n emitting layer, where data points with $|\cos(\Delta \theta)| < 0.7$ are excluded.
- Fig. S17 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 1
- Fig. S18 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of 1

Fig. S19	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of 2
Fig. S20	¹³ C{ ¹ H} NMR spectrum (126 MHz, CD ₂ Cl ₂) of 2
Fig. S21	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of 3
Fig. S22	$^{13}\text{C}\{^{1}\text{H}\}$ NMR spectrum (126 MHz, CD ₂ Cl ₂) of $\boldsymbol{3}$
Fig. S23	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of 4
Fig. S24	$^{13}\text{C}\{^{1}\text{H}\}$ NMR spectrum (126 MHz, CD ₂ Cl ₂) of $\textbf{4}$
Fig. S25	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of 5
Fig. S26	$^{13}\text{C}\{^{1}\text{H}\}$ NMR spectrum (126 MHz, CD ₂ Cl ₂) of $\textbf{5}$
Fig. S27	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of (S)-6
Fig. S28	¹³ C{ ¹ H} NMR spectrum (126 MHz, CD ₂ Cl ₂) of (S)-6
Fig. S29	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of (<i>R</i>)-6
Fig. S30	¹³ C{ ¹ H} NMR spectrum (126 MHz, CD ₂ Cl ₂) of (<i>R</i>)-6
Fig. S31	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of (S)-Pt
Fig. S32	¹³ C{ ¹ H} NMR spectrum (126 MHz, CD ₂ Cl ₂) of (S)-Pt
Fig. S33	¹ H NMR spectrum (300 MHz, CD ₂ Cl ₂) of (<i>R</i>)-Pt
Fig. S34	¹³ C{ ¹ H} NMR spectrum (126 MHz, CD ₂ Cl ₂) of (<i>R</i>)-Pt
References	

Experimental Details

Materials and General Methods. The synthesis of $[Pt_2(\mu-Cl)_2(2-phenylpyridinate)_2]$ has been reported previously.¹ Commercially available chemicals were purchased and used without further purification. (R)-(-)-1-Phenyl-2-propanol (>97.0%), phosphorus(V) oxychloride (99.0%), methyltriphenylphosphonium bromide (98.0%), pyridine (99.8%), dimethyl sulfoxide (>99.9%), N,N-dimethylformamide (99.8%), 1,4-dioxane (>99.0%), 1-methyl-2-pyrrolidinone (99.5%), 2-(tributylstannyl)pyridine (85%), toluene (99.8%), tetrakis(triphenylphosphine)palladium(0) (99%), and 2-ethoxyethanol (99%) were purchased from Sigma Aldrich. p-Toluenesulfonyl chloride (>99.0%), carbazole (>97.0%), 18-crown-6 ether (>98.0%), N-ethylcarbazole-3-carboxaldehyde (>98.0%), (R)-3,3'-dibromo-1,1'-bi-2-naphthol (>98%), and (S)-3,3'-dibromo-1,1'-bi-2-naphthol (>97%) were purchased from Tokyo Chemical Industry. Potassium hydroxide (>93.0%), potassium carbonate (>99.5%), and 2,2-azobisisobutyronitrile (>99.0%) were purchased from Daejung Chemicals. All glassware and magnetic stirring bars were thoroughly dried in a convection oven. The reactions were monitored by using thin layer chromatography (TLC). Commercial TLC plates (silica gel 60 F254, Merck Co.) were developed and the spots were visualized under UV illumination at 254 or 365 nm. Silica gel column chromatography was performed by using silica gel 60 (particle size 0.063-0.200 mm, Merck Co.). ¹H and ¹³C[¹H] NMR spectra were collected with Bruker, Ultrashield 500 or 300 plus NMR spectrometers. Chemical shifts were referenced to (CH₃)₄Si. High resolution mass spectra (positive mode, FAB) were obtained by employing a JEOL, JMS-600W mass spectrometer.

Synthesis of 1. (*R*)-(–)-1-Phenyl-2-propanol (10.4 g, 76.4 mmol) and *p*-toluenesulfonyl chloride (16.0 g, 83.7 mmol) were dissolved in pyridine (40 mL) in a 250 mL one-necked round-bottom flask equipped with a magnetic stir bar. The mixture was stirred at room temperature for 1 day under an Ar atmosphere. The reaction mixture was poured onto EtOAc (400 mL) and washed thoroughly with water (400 mL × three times). The organic layer was recovered, dried over anhydrous MgSO₄ and concentrated under a reduced pressure. Silica gel column chromatography was performed with increasing the polarity of the eluent from EtOAc:hexane = 1:9 (v/v) to EtOAc:hexane = 1:1 (v/v). A white powder was obtained in a 94% yield. *R*_f = 0.29 (EtOAc:hexane = 1:4, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 1.28 (d, *J* = 6.3 Hz, 3H), 2.41 (s, 3H), 2.74–2.90 (m, 2H), 4.69 (sextet, *J* = 6.3 Hz, 1H), 7.01–7.05 (m, 2H), 7.17–7.26 (m, 5H), 7.58 (d, *J* = 9.0 Hz, 2H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 20.9, 21.9, 43.3, 81.3, 127.1, 128.0, 128.9, 130.0, 130.2, 134.4, 137.0, 145.1. HR MS (FAB, *m*-NBA): Calcd for C₁₆H₁₈O₃S ([M+H]⁺), 291.1050; found: 291.1053.

Synthesis of 2. Potassium carbonate (5.70 g, 102 mmol), carbazole (12.7 g, 76.1 mmol), and 18crown-6 (25.2 g, 95.3 mmol) were added into a 250 mL two-necked round-bottom flask. The reagents were dissolved in DMSO (50 mL), and the mixture was heated at 80 °C for 1 h. Compound **1** (14.8 g, 50.8 mmol) dissolved in 30 mL of DMSO was added to the stirred solution, which was stirred for additional 2 days. After cooling the reaction mixture to room temperature, it was poured onto EtOAc (400 mL) and washed thoroughly with water (400 mL × three times). The organic layer was recovered, dried over anhydrous MgSO₄ and concentrated in vacuo. Purification by silica gel column chromatography was performed with increasing the polarity of the eluent from CH₂Cl₂:hexane = 1:19 (v/v) to CH₂Cl₂:hexane = 1:1 (v/v). A pale yellow solid was obtained in a 7% yield. *R*₁ = 0.50 (CH₂Cl₂:hexane = 1:2, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 1.74 (d, *J* = 6.9 Hz, 3H), 3.26–3.53 (m, 2H), 5.00 (sextet, *J* = 6.9 Hz, 1H), 7.01–7.21 (m, 7H), 7.37–7.42 (m, 4H), 8.07 (d, *J* = 7.8 Hz, 2H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 19.2, 41.5, 110.6, 119.1, 120.7, 125.9, 126.9, 128.8, 129.4, 139.5. HR MS (FAB, *m*-NBA): Calcd for C₂₁H₁₉N ([M]⁺), 285.1517; found: 285.1514.

Synthesis of 3. *N*,*N*-Dimethylformamide (1.53 g, 20.9 mmol) and phosphorus(V) oxychloride (2.15 g, 14.0 mmol) were placed in a 25 mL two-necked round-bottom flask. The mixture was stirred for 1 h at 0 °C under an Ar atmosphere. Compound **2** (1.00 g, 3.50 mmol) dissolved in 8 mL DMF was slowly added to the stirred solution at 0 °C, by using a syringe. The mixture was heated then at 110 °C for 1 day. After cooled the reaction mixture to room temperature, it was poured onto EtOAc (200 mL) and washed thoroughly with water (200 mL × five times). The recovered organic layer was dried over anhydrous MgSO₄, filtered, and concentrated under a reduced pressure. Silica gel column chromatography was performed with CH₂Cl₂:hexane = 1:1 (v/v) as an eluent. The purified compound was obtained as a yellow solid (66%). *R*_f = 0.20 (CH₂Cl₂:hexane = 1:1, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 1.79 (d, *J* = 7.2 Hz, 3H), 3.28–3.54 (m, 2H), 5.05 (sextet, *J* = 7.2 Hz, 1H), 6.97–3.99 (m, 2H), 7.05–7.08 (m, 3H), 7.27–7.31 (m, 1H), 7.48–7.50 (m, 3H), 7.89–7.95 (m, 1H), 8.15 (d, *J* = 7.8 Hz, 1H), 8.57 (s, 1H), 10.05 (s, 1H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 19.3, 30.3, 41.4, 109.7, 112.5, 120.6, 121.1, 124.0, 126.9, 127.0,128.8, 128.9, 129.2, 138.9, 191.9. HR MS (FAB, *m*-NBA): Calcd for C₂₂H₁₉NO ([M+H]⁺), 314.1540; found: 314.1548.

Synthesis of 4. Potassium carbonate (13.8 g, 99.6 mmol) and methyltriphosphonium bromide (35.7 g, 99.8 mmol) were dissolved in 1,4-dioxane (50 mL) in a 250 mL two-necked round-bottom flask equipped with a magnetic stir bar. Compound **3** (1.3 g, 4.15 mmol) dissolved in 34 mL of 1,4-dioxane was delivered into the reaction mixture, then milli-Q water (7 mL) was added. The resulting

mixture was stirred at 110 °C for 4 days under an Ar atmosphere. After cooling the reaction mixture to room temperature, it was poured onto EtOAc (400 mL) and thoroughly washed with water (400 mL × three times). The organic layer was dried over anhydrous MgSO₄, filtered, and concentrated under a reduced pressure. Silica gel column chromatography was performed with increasing the polarity of the eluent from CH₂Cl₂:hexane = 1:49 (v/v) to CH₂Cl₂:hexane = 1:1 (v/v). A yellow viscous solid was obtained in a 40% yield. R_f = 0.63 (CH₂Cl₂:hexane = 1:1, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 1.74 (d, *J* = 6.9 Hz, 3H), 3.25–3.50 (m, 2H), 4.82 (sextet, *J* = 6.9 Hz, 1H), 5.18 (d, *J* = 10.8 Hz, 1H), 5.77 (d, *J* = 17.4 Hz, 1H), 6.89 (dd, *J* = 10.8, 17.6 Hz, 1H), 7.00–7.16 (m, 5H), 7.18–7.21 (m, 1H), 7.37–7.52 (m, 4H), 8.05–8.09 (m 2H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 19.0, 30.3, 41.4, 110.2, 118.7, 120.6, 123.7, 125.2, 126.0, 126.6, 128.6, 129.3, 136.4, 138.2, 139.5, 140.4. HR MS (FAB, *m*-NBA): Calcd for C₂₃H₂₁N ([M]⁺), 311.1674; found: 311.1674.

Synthesis of 5. Compound **5** was prepared with the method used for the synthesis of compound **4**, except the use of *N*-ethylcarbazole-3-carboxaldehyde in place of compound **3**. A white solid was obtained in a 55% yield. $R_f = 0.63$ (CH₂Cl₂:hexane = 1:1, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 1.42 (t, J = 7.2 Hz, 3H), 4.37 (q, J = 7.2 Hz, 2H), 5.19 (d, J = 10.8 Hz, 1H), 5.78 (d, J = 17.7 Hz, 1H), 6.92 (dd, J = 17.6, 10.9 Hz, 1H), 7.20–7.25 (m, 1H), 7.38–7.50 (m, 3H), 7.59 (d, J = 8.4 Hz, 1H), 8.08–8.11 (m, 2H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 14.1, 30.3, 108.6, 118.7, 119.9, 120.7, 122.7, 123.4, 125.4, 136.8, 138.7, 138.9, 140.4, 140.5. HR MS (FAB, *m*-NBA): Calcd for C₁₆H₁₅N ([M]⁺), 221.1204; found: 221.1206.

Synthesis of (S)-6. (S)-3,3'-Dibromo-1,1'-bi-2-naphthol (0.958 g, 2.16 mmol) and tetrakis(triphenylphosphine)palladium(0) (0.251 g, 0.217 mmol) were dissolved in anhydrous toluene (50 mL) in a 100 mL two-necked round-bottom flask equipped with a magnetic stir bar. 2- (Tributylstannyl)pyridine (2.00 g, 5.43 mmol) dissolved in 2 mL of anhydrous toluene was delivered into a stirred solution under an Ar atmosphere. The solution was stirred at 115 °C for 21 h. After cooling the reaction mixture to room temperature, it was concentrated under a reduced pressure. The concentrate was subjected to silica gel column chromatography with increasing the polarity of the eluent from CH₂Cl₂:hexane = 3:1 (v/v) to CH₂Cl₂:hexane = 7:1 (v/v). A yellow powder was obtained in a 14% yield. *R*_f = 0.48 (CH₂Cl₂). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 5.25 (s, 1H), 7.07 (d, *J* = 16.5 Hz, 1H), 7.15 (d, *J* = 14.4 Hz, 1H), 7.23 (dd, *J* = 14.4, 5.6 Hz, 1H), 7.29–7.34 (m, 4H), 7.89 (d, *J* = 8.1 Hz, 1H), 7.93 (d, *J* = 8.7 Hz, 1H), 8.00–8.02 (m, 2H), 8.31 (d, *J* = 8.4 Hz, 1H), 8.53 (d, *J* = 4.8 Hz, 1H), 8.62 (s, 1H), 14.7 (s, 1H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 114.7, 116.1, 118.2, 120.9, 122.1, 123.2, 123.8, 124.2, 124.6, 125.3, 126.9, 128.4, 128.7, 128.71, 128.9,

129.6, 129.8, 130.2, 134.3, 135.7, 139.0, 146.5, 152.1, 156.5, 157.8. HR MS (FAB, *m*-NBA): Calcd for C₂₅H₁₆BrNO₂ ([M–Br+H]⁺), 364.1332; found: 364.1331.

Synthesis of (*R***)-6. (***R***)-6 was prepared following the method identical to the synthesis of the compound (***S***)-6, except the use of (***R***)-3,3'-dibromo-1,1'-bi-2-naphthol in place of (***S***)-3,3'-dibromo-1,1'-bi-2-naphthol. A yellow powder was obtained in a 15% yield. R_f = 0.48 (CH₂Cl₂). ¹H NMR (300 MHz, CD₂Cl₂) \delta (ppm): 5.28 (s, 1H), 7.10 (d,** *J* **= 16.8 Hz, 1H), 7.19 (d,** *J* **= 15.9 Hz, 1H), 7.27 (dd,** *J* **= 14.5, 5.7 Hz, 1H), 7.29–7.34 (m, 4H), 7.92 (d,** *J* **= 8.1 Hz, 1H), 7.96 (d,** *J* **= 8.7 Hz, 1H), 8.03–8.05 (m, 2H), 8.34 (d,** *J* **= 8.4 Hz, 1H), 8.57 (d,** *J* **= 4.8 Hz, 1H), 8.66 (s, 1H), 14.8 (s, 1H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) \delta (ppm): 114.7, 116.0, 118.2, 120.9, 122.1, 123.2, 123.8, 124.2, 124.6, 125.3, 126.9, 128.4, 128.7, 128.71, 128.9, 129.6, 129.8, 130.2, 134.3, 135.7, 139.0, 146.5, 152.1, 156.4, 157.8. HR MS (FAB,** *m***-NBA): Calcd for C₂₅H₁₆BrNO₂ ([M(–Br)+H]⁺), 364.1333; found: 364.1340.**

Synthesis of (S)-Pt. (S)-6, $[Pt_2(\mu-Cl)_2(2-phenylpyridinate)_2]$, and potassium carbonate were dissolved in 2-ethoxyethanol (25 mL) in a 50 mL one-necked round-bottom flask equipped with a magnetic stir bar. The mixture was stirred at 60 °C for 16 h under an Ar atmosphere. After cooling the reaction mixture to room temperature, it was poured onto water (400 mL) and then neutralized with 1 N HCl(aq). The crude product was extracted with CH₂Cl₂ (400 mL × three times). The organic layer was dried over anhydrous MgSO₄, filtered, and concentrated in vacuo. Silica gel column chromatography was performed with increasing the polarity of the eluent from CH₂Cl₂:CH₃OH = 99:1 (v/v) to CH₂Cl₂:CH₃OH = 49:1 (v/v). The purified compound was obtained as an orange powder (90%). R_f = 0.54 (CH₂Cl₂:CH₃OH = 19:1, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 6.94–7.08 (m, 5H), 7.14–7.23 (m, 4H), 7.36–7.51 (m, 5H), 7.68 (broad m, 1H), 7.85–7.89 (m, 2H), 7.92 (d, *J* = 9.0 Hz, 1H), 8.03 (d, *J* = 6.9 Hz, 1H), 8.13 (d, *J* = 7.8 Hz, 1H), 8.19 (s, 1H), 9.11 (d, *J* = 4.5 Hz, 1H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 118.2, 119.2, 119.6, 121.3, 122.3, 123.6, 123.8, 124.3, 124.6, 126.1, 126.4, 126.5, 127.5, 127.9, 128.5, 129.3, 129.4, 129.8, 129.9, 131.0, 132.8, 133.5, 134.9, 137.4, 138.9, 1339.5, 140.5, 146.4, 148.2, 153.1, 154.8, 157.8, 166.8. HR MS (FAB, *m*-NBA): Calcd for C₃₆H₂₃BrN₂O₂Pt ([M(–Br])⁺), 712.1558; found: 712.1585.

Synthesis of (*R***)-Pt. (***R***)-Pt** was prepared following the method identical to the synthesis of the compound (*S*)-Pt, except the use of compound (*R*)-6 in place of (*S*)-6. An orange powder was obtained in a 96% yield. $R_f = 0.54$ (CH₂Cl₂:CH₃OH = 19:1, v/v). ¹H NMR (300 MHz, CD₂Cl₂) δ (ppm): 6.94–7.08 (m, 5H), 7.14–7.23 (m, 4H), 7.36–7.45 (m, 5H), 7.68 (broad m, 1H), 7.85–7.89 (m, 2H), 7.98 (d, *J* = 9.0 Hz, 1H), 8.02 (d, *J* = 7.2 Hz, 1H), 8.13 (d, *J* = 8.5 Hz, 1H), 8.19 (s, 1H), 9.11 (d, *J* = 6.0 Hz, 1H). ¹³C[¹H] NMR (126 MHz, CD₂Cl₂) δ (ppm): 118.2, 119.2, 119.6, 121.3,

122.3, 123.5, 123.8, 124.3, 124.6, 126.1, 126.4, 126.5, 127.5, 127.9, 128.5, 129.3, 129.4, 129.8, 129.9, 131.0, 132.8, 133.5, 134.9, 137.4, 138.9, 1339.5, 140.5, 146.4, 148.2, 153.1, 154.8, 157.8, 166.8. HR MS (FAB, *m*-NBA): Calcd for C₃₆H₂₃BrN₂O₂Pt ([M(–Br)]⁺), 712.1558; found: 712.1599.

Free-Radical Copolymerization. The copolymers JY_n were prepared by performing free radical polymerizations of chiral (**4**) and achiral monomers (**5**) in the presence of AIBN as a thermal initiator. In the case of JY_1 , **4** (8.60 mg, 0.028 mmol), **5** (608 mg, 2.76 mmol), and AIBN (9.16 mg, 55.8 mmol) were delivered into a 4 mL glass ampoule containing a magnetic stir bar. After the addition of 2.5 mL of anhydrous 1-methyl-2-pyrrolidinone into the ampoule, the solution was thoroughly degassed by carrying out repeated vacuum–freeze–thaw cycles. The ampoule was sealed with a torch. The polymerization was performed by heating the solution at 65 °C for 2 days under dark conditions. After cooling the reaction mixture to room temperature, it was poured into 200 mL of stirred methanol to form a white precipitate. The precipitate was filtered and thoroughly washed with methanol, and the filter cake was re-dissolved in CH₂Cl₂. After concentration, the reprecipitation and dissolution cycle was repeated. The collected polymers were dried in a vacuum oven.

Polymer Characterizations. Differential scanning calorimetry (DSC) was performed using a TA Instruments, DSC 250 at a heating rate of 10 °C min⁻¹ under a nitrogen atmosphere. Calorimetric analyses were performed by using the software provided by the manufacturers. Thermogravimetric analysis (TGA) was performed on a Mettler Toledo, TGA/DSC 3+ instrument at a heating rate of 10 °C min⁻¹ under a nitrogen atmosphere. Typically, $T_{d,90}$, which is the temperature at which the weight reached 90% of the original value, was determined. Gel permeation chromatography (GPC) was performed on a Thermo Scientific, Ultimate 3000 instrument equipped with an LPG-3400SD quaternary pump, a Refracto Max 520 refractive index detector, a Shodex KF-806L column, a Shodex KF-G guard column, and a Shodex standard SM-105. THF was used as the eluent, and the flow rate was 1.0 mL min⁻¹.

Steady-State UV–vis Absorption Spectroscopy. UV–vis absorption spectra were collected on an Agilent, Cary 300 spectrophotometer at 298 K. Sample solutions were prepared prior to measurements at a concentration of 10 μ M in CH₂Cl₂ or 1,2-dichloroethane, unless otherwise stated. Each solution was delivered into a quartz cell (Hellma, beam path length = 1.0 cm). Neat films of the JY_n polymers were prepared by dissolving 5 mg JY_n in 245 mg 1,2-dichloroethane (i.e., 2.0 wt % total solute). The solution was spincoated onto a 2.5 cm × 2.5 cm quartz plate at 2000 rpm. JY_n films doped with (*R*)- and (*S*)-Pt were prepared by using 1,2-dichloroethane (245–500 mg) containing 0–0.5 mg Pt dopants and 4.5–10.0 mg JY_n (i.e., 2.0 wt % total solute; Pt: JY_n = 0:100-10:90, w/w).

Electronic Circular Dichroism Measurements. Electronic circular dichroism (ECD) spectra were collected on a Jasco, J1500 spectropolarimter at 298 K under a nitrogen atmosphere. The neat films prepared for UV–vis absorption spectroscopy analysis were used in the ECD measurements. The measurements were repeated three times for each sample at various positions. Measurement conditions: scan rate, 200 nm min⁻¹; detection range, 200–600 nm.

Steady-State Photoluminescence Measurements. Photoluminescence spectra were obtained by using a Photon Technology International, Quanta Master 400 scanning spectrofluorometer at 298 K. The solutions (1,2-dichloroethane) and films (quartz plates) used in the steady-state UV–vis absorption studies were employed in the photoluminescence measurements. The solutions of (*R*)and (*S*)-Pt were deaerated by bubbling Ar through them for 15 min prior to the measurements. A quartz cell (Hellma, beam path length = 1.0 cm) was employed for the solution samples. The excitation wavelengths were 400 nm for (*R*)- and (*S*)-Pt, and 339 nm for JY_n. The spectra were recorded in the emission range 350–800 nm.

Determination of Photoluminescence Lifetimes. Deaerated 1,2-dichloroethane solutions of 100 μ M (*R*)- and (*S*)-Pt were employed. The films were freshly prepared, prior to the measurements. Photoluminescence decay traces were acquired based on time-correlated single-photon-counting (TCSPC) techniques, by using a PicoQuant, FluoTime 200 instrument after nanosecond pulsed laser excitation at 377 nm for JY_n and at 332 nm for (*R*)- and (*S*)-Pt (pulse duration = 25 ps). Transient photon signals were collected at λ_{obs} = 420 nm (JY_n) or at 590 nm ((*R*)- and (*S*)-Pt) through an automated motorized monochromator. The photon acquisition was terminated when the accumulated photon count reached 10⁴. Photoluminescence decay traces were analyzed by using the mono- or triexponential decay models embedded in an OriginLab, OriginPro 2018 software. In the case of triphasic decay, average photoluminescence lifetime (τ_{obs}) values were calculated by using the relationship $\tau_{obs} = \Sigma A_i \pi^2 / \Sigma A_i \pi$ (*i* = 1–3), where A_i and π are the pre-exponential factor and the time constant, respectively.

Determination of Relative Photoluminescence Quantum Yields. Photoluminescence quantum yields (PLQYs) of solution samples were determined relatively, by using the equation PLQY = $PLQY_{ref} \times (I/I_{ref}) \times (A_{ref}/A) \times (n/n_{ref})^2$, where *A*, *I*, and *n* are the absorbance at the excitation wavelength, the integrated photoluminescence intensity, and the refractive index of the solvent, respectively. 9,10-Diphenylanthracene (PLQY = 1.00, toluene; λ_{ex} = 366 nm) was used as the

reference material.² Typically, 10 μ M samples were dissolved in 1,2-dichloroethane which that had been thoroughly deaerated prior to the measurements. Photoluminescence spectra were collected at 298 K in the emission range 400–800 nm. The spectra were integrated with an OriginLab, OriginPro 2019 software.

Determination of Absolute Photoluminescence Quantum Yields. Photoluminescence quantum yields (PLQYs) of films were determined with an absolute method. Films were freshly prepared on 2.5 cm \times 2.5 cm quartz plates prior to the measurements. The photon flux of each excitation beam (330 nm for non-doped JY_n films; 353 nm for Pt-doped JY_n films; $l_{ex}(0)$) was quantified in the absence of a film. The film was placed into an integrating sphere (Photon Technology International), and the excitation beam was focused at the center of the sample. The absorption of the excitation photons by each film ($l_{ex}(s)$) was quantified. Finally, the photon flux of the photoluminescence emission from each sample (l_{em}) was measured under the following conditions: integration time, 0.1 s; step size, 0.0625 nm; emission ranges, 300–650 nm for the JY_n films and 330–700 nm for the Pt-doped JY_n films. PLQY was calculated by using the relationship, PLQY = $l_{em}/(l_{ex}(s))$. The measurements were repeated in triplicate for each fresh sample.

Circularly Polarized Luminescence Measurements. CPL measurements were performed using a home-made setup shown in ESI, Fig. S14. Light emitted by a sample was modulated by a photoelastic modulator (PEM, PEM-100, Hinds Instruments) providing sinusoidal guarter-wave retardation oscillating at 47 kHz, and then was passed through a linear polarizer oriented at 45° with respect to the PEM axis. The optical power detected by a photomultiplier tube (PMT, R9182-01, Hamamatsu) was amplified with a current amplifier (C11184, Hamamatsu). The DC and AC components of the amplified signal were measured using a digital multimeter (34410A, Agilent) and a lock-in amplifier (SR830, Stanford Research Systems), respectively. The operation of our CPL spectroscopic system was confirmed by reproducing the gPL spectrum of 5.5 mM europium tris[3-(trifluoromethylhydroxymethylene)-(+)-camphorate] ([Eu(facam)₃]) diluted in DMSO.³ For the g_{PL} measurements, samples were excited by an LED with a peak wavelength of 365 nm (M365LP1, Thorlabs), while for the g_{EL} measurements, PLEDs were packaged in a N₂-filled glovebox before the measurements and were driven by a source measure unit (2400 SourceMeter, Keithley). The full width at half maximum resolution of the spectrometer was set to be 13 and 19 nm for the g_{PL} and g_{EL} measurements, respectively. Small values of both g_{EL} and the intensity of the PLED emission limited the signal-to-noise ratio in the gEL measurements, leading to the phase difference $\Delta \theta$ between the sinusoidal reference signal produced by the PEM and the output of the current amplifier deviating from 0 or π , as shown in ESI, Fig. S15. Based on the assumption that the data

points with $\Delta\theta$ deviating far from 0 or π are less reliable than others, we also show the g_{EL} spectra where the data points with $|\cos(\Delta\theta)| < 0.7$ are excluded (ESI, Fig. S16).

Confocal Laser Scanning Microscopy. Films of (*S*)-Pt/JY_n were freshly prepared by spincoating 1,2-dichloroethane solutions containing various ratios of (*S*)-Pt and JY_n (2.0 wt % total solute; (*S*)-Pt:JY_n = 10:90–50:50, w/w) onto 2.5 cm × 2.5 cm quartz plates. Photoluminescence images of the films were visualized by employing a Carl Zeiss, LSM780 NLO confocal laser scanning microscope. An excitation beam (405 nm) was focused onto the film, and the photoluminescence signals were acquired for the emission range 500–700 nm. The acquired images were analyzed by using Carl Zeiss, ZEN 2.3 SP1 version 14.0 software.

Transmission Electron Microscopy. Films of (*S*)-Pt/JY_n were freshly prepared by spincoating 1,2-dichloroethane solutions (1.0 wt % total solute; (*S*)-Pt:JY_n = 50:50, w/w) onto 200-mesh carbon films on a copper grid. TEM images were acquired by employing a JEOL, JEM-2100F. The atomic compositions were quantified for Cliff Lorimer thin ratio sections of the samples by using energy dispersive spectroscopy.

Electrochemical Characterization. Cyclic and differential pulse voltammetry experiments were carried out by using a CH Instruments, CHI630 B potentiostat with a three-electrode-cell assembly. A Pt wire and a Pt microdisc were used as the counter and working electrodes, respectively. A Ag/AgNO₃ couple was used as a pseudo reference electrode. Measurements were carried out in Ar-saturated THF (2.0 mL) by using 0.10 M tetra-*n*-butylammonium hexafluorophosphate (Bu₄NPF₆) as the supporting electrolyte at scan rates of 100 mV s-1 (cyclic voltammetry) or 4.0 mV s-1 (differential pulse voltammetry). A ferrocenium/ferrocene couple was employed as the external reference.

Device Fabrication and Characterization Glass substrates precoated with an ITO anode were sequentially cleaned in ultrasonic baths of detergent, de-ionized water, acetone, and isopropyl alcohol, followed by UV–ozone treatment for 30 min. A GraHIL layer was deposited by spin-coating (4500 rpm, 60 s) a solution of PEDOT:PSS (P VP AI 4083, CLEVIOS) and tetrafluoroethylene-perfluoro-3,6-dioxa-4-methyl-7-octenesulphonic acid copolymer (Nafion, DuPont), followed by a 30-min bake on a hot plate at 150 °C. The Pt:JY_n solution was prepared by dissolving the polymer host and the Pt dopant at a doping concentration of 30 wt % in 1,2-dichloroethane (0.5 wt % total solute), which was filtered using a 0.45-µm pore-size PTFE syringe filter (Whatman) before spin-coating (5000 rpm, 40 s) on a GraHIL layer in a N₂-filled glovebox. After a 30-min bake on a hot plate at 100 °C, the samples were loaded into a vacuum chamber (~10⁻⁷ torr), where layers of

TPBi (50 nm), LiF (1 nm), and Al (100 nm) were sequentially deposited by thermal evaporation. The current density–voltage–luminance characteristics were obtained using a source measure unit (2400 SourceMeter, Keithley) and a Si photodiode (818-SL, Newport). The electroluminescence spectra were obtained using a spectrometer (SR303i, Shamrock) coupled with a CCD camera (iDus 401, Andor).



Fig. S1 Photoluminescence decay traces for thin films of JY_n polymers spincast on quartz substrates recorded at wavelengths of 380 nm (a) and 420 nm (b) after pulsed laser photoexcitation of 332 nm (temporal resolution = 25 ps). The 380 and 420 nm emissions correspond to the fluorescence emission of half-sandwich and full-sandwich excimers, respectively. Instrumental response function (IRF) is included. The rise profiles of the 420 nm traces are distinguishable from IRF. The solid curves in (b) are nonlinear least-squares fits of the traces to an exponential rise and decay model. The rise time (τ_{rise}) of JY₀ is 0.52 ns, and τ_{rise} of JY₁₀₀ is 0.56 ns.



Fig. S2 (a) Comparisons of the UV–vis absorption (top), ECD (middle), and the Kuhn dissymmetry factor (g_{abs}) spectra of a 100 µM solution of **2** (black curves) and a 0.085 mg mL⁻¹ solution of JY₁₀₀ (red curves) in 1,2-dichloroethane. The silence of **2** implies the essential role of the asymmetric exciton coupling among the neighboring carbazole pendants for the ECD signaling. (b) Comparisons of the UV–vis absorption (top), ECD (middle), and the Kuhn dissymmetry factor (g_{abs}) spectra of JY₁₀₀ solutions obtained with increased concentrations (0.017–0.17 mg mL⁻¹ in 1,2-dichloroethane). The invariance of the g_{abs} plots indicates that the ECD signaling results from the intrachain exciton coupling of carbazole pendants.



Fig. S3 ECD spectra of a 0.028 mg mL⁻¹ solution of JY₁₀₀ (1,2-dichloroethane) recorded upon cooling (a) and heating (b). The temperature control was achieved using a Peltier heater equipped in a Jasco, J1500 spectropolarimter. The ellipticity decreases with temperature, which indicates that the extent of asymmetric exciton coupling decreases at elevated temperature due to thermally activated increases in the intrachain conformational freedom.



Fig. S4 Plots of the Kuhn dissymmetry factor (g_{abs}) as a function of wavelength for dropcast films of *N*-ethylcarbazole doped with varying concentrations of **2** (0.5–15 wt %). The negligible g_{abs} values of the films imply that the ECD signaling of the JY_n films originates from intrachain exciton coupling among neighboring carbazole pendants.



Fig. S5 ECD spectra of the neat films of JY_n polymers cast on quartz substrates.



Fig. S6 (a) Plots of the Kuhn dissymmetry factor (g_{abs}) as a function of wavelength for the neat films of the JY_n polymers doped with 10 wt % (R)-Pt. The inset arrow indicates that g_{abs} of the MLCT transition band increases with *n* of the JY_n polymers. (b) Plot of g_{abs} ($\lambda_{obs} = 372$ nm) as a function of the mole fraction of the chiral repeat unit (i.e., [M*] / ([M*] + [M])) in JY_n. The non-linear increase of g_{abs} indicates amplification of chiroptical properties due to the chiral repeat unit in the JY polymers.



Fig. S7 (a) UV–vis absorption (top) and ECD (bottom) spectra of 10 μ M solutions of (*R*)- and (*S*)-Pt (1,2-dichloroethane). (b) Normalized photoluminescence spectra of 10 μ M solutions of (*R*)- and (*S*)-Pt (1,2-dichloroethane) recorded upon photoexcitation at 400 nm.



Fig. S8 g_{PL} spectra of a 100 mM (*R*)-Pt in 1,2-dichloroethane (1,2-DCE; red symbols) and JY₀ (empty blue symbols) and JY₁₀₀ (filled blue symbols) films doped with 30 wt % (*R*)-Pt.



Fig. S9 (a,b) Photoluminescence spectra ($\lambda_{ex} = 339$ nm) obtained for JY₀ (a) and JY₁₀₀ (b) films doped with various concentrations (0–10 wt %) of (*S*)-Pt. (c,d) Photoluminescence decay traces ($\lambda_{obs} = 420$ nm) of JY₀ (c) and JY₁₀₀ (d) films doped with various concentrations (0–10 wt %) of (*S*)-Pt obtained after picosecond pulsed laser photoexcitation at 377 nm (temporal resolution = 25 ps). The solid curves correspond to nonlinear least-squares fits to a biexponential decay model.



Fig. S10 Photoluminescence decay traces of 10 wt % (*S*)-Pt:JY₀ films (a) and 10 wt % (*S*)-Pt:JY₁₀₀ films observed at wavelengths of 420 (filled symbols; i.e., host emission) and 590 nm (empty symboles; i.e., dopant emission) after picosecond pulsed laser photoexcitation at 377 nm. Instrumental response function (IRF) obtained using a non-emissive scattering medium is included for comparison (black symbols).



Fig. S11 Cyclic (solid curve) and differential pulse (dotted line) voltammograms of a 0.39 mg mL⁻¹ solution of PVK (Ar-saturated THF) containing 0.10 M Bu₄NPF₆ supporting electrolyte. A Pt disk and a Pt wire for the working and counter electrodes, respectively; the Ag/AgNO₃ (10 mM) pseudo reference electrode; scan rate = 0.10 V s⁻¹ (CV) and 4 mV s⁻¹ (DPV).



Fig. S12 Electroluminescence spectra of the PLEDs employing (*R*)-Pt:JY₀ with different doping concentrations of (*R*)-Pt (10, 20, and 30 wt %) as emitting layers. Our choice of the doping concentration (30 wt %) for the results shown in Fig. 9 was based on the fact that the (*R*)-Pt:JY₀ PLED with 30 wt % doping has the smallest host emission in the short wavelength region (< 500 nm). However, devices with 10 and 20 wt % doping are likely to yield the g_{EL} values comparable to those shown in Fig. 9, because the host emission from the (*R*)-Pt:JY₀ PLEDs is negligible in the wavelength region where the g_{EL} measurements were performed as shown in this plot.



Fig. S13 g_{PL} spectra of the 30 wt %-doped films measured with the substrates facing the PEM, showing that the signs of g_{PL} are the same as those of the measurements with the sample films facing the PEM (Fig. 5 in the main text).



Fig. S14 Schematic diagram of the home-made CPL spectroscopic system.



Fig. S15 g_{EL} spectra and $\cos(\Delta\theta)$ plots of the PLEDs with a (*R*)- or (*S*)-Pt:JY_n emitting layer, showing all data points used to plot Fig. 9e. Solid symbols represent the averages of 20 repeated measurements. The signs of g_{EL} are determined to be positive (or negative) when $\cos(\Delta\theta) > 0$ (or < 0).



Fig. S16 g_{EL} spectra and $\cos(\Delta \theta)$ plots of the PLEDs with a (*R*)- or (*S*)-Pt:JY_n emitting layer, where data points with $|\cos(\Delta \theta)| < 0.7$ are excluded.



Fig. S17 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 1.



Fig. S18 $^{13}\text{C}\{^{1}\text{H}\}$ NMR spectrum (126 MHz, CD_2Cl_2) of 1.



Fig. S19 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 2.



Fig. S20 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of 2.



Fig. S21 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 3.



Fig. S22 ${}^{13}C{}^{1}H$ NMR spectrum (126 MHz, CD₂Cl₂) of 3.



Fig. S23 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 4.



Fig. S24 ${}^{13}C{}^{1}H$ NMR spectrum (126 MHz, CD₂Cl₂) of 4.



Fig. S25 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of 5.



Fig. S26 $^{13}C\{^{1}H\}$ NMR spectrum (126 MHz, CD₂Cl₂) of 5.



Fig. S27 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of (S)-6.



Fig. S28 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of (S)-6.



Fig. S29 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of (*R*)-6.



Fig. S30 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of (*R*)-6.



Fig. S31 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of (S)-Pt.



Fig. S32 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of (S)-Pt.



Fig. S33 ¹H NMR spectrum (300 MHz, CD₂Cl₂) of (*R*)-Pt.



Fig. S34 ¹³C{¹H} NMR spectrum (126 MHz, CD₂Cl₂) of (*R*)-Pt.

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