Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Electronic Supplementary Information

Circularly polarized electroluminescence by controlling emission zone in twisted liquid crystalline conjugate polymer

Jin-Hyung Jung, Dong-Myung Lee, Jae-Hoon Kim and Chang-Jae Yu*

Department of Electronic Engineering, Hanyang University, Seoul 04763, Korea.

**E-mail: cjyu@hanyang.ac.kr*

Evaluation the Total Twist Angle

For the evaluation of total twist angle of the F8BT layer with 120 nm thickness, the Stokes parameters, passing through the continuously twisted F8BT layer of the incident probing light with linear polarization, were measured and compared to the theoretical parameters as shown in Fig. S1. All measured Stokes parameters were depicted by horizontal lines and the calculated parameters based on the Jones matrix analysis for the twisted birefringent media were presented by open symbols.[1] Three Stokes parameters were matched at 86°.

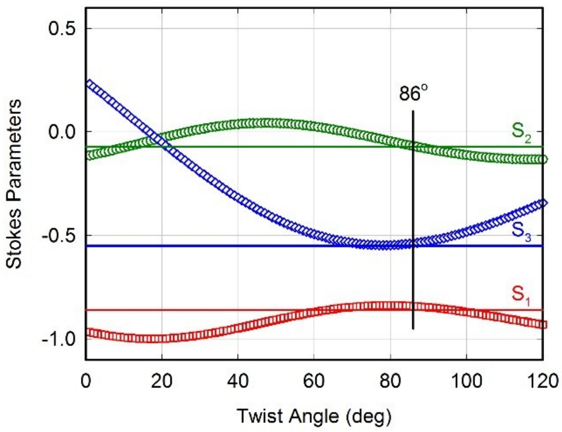

Fig. S1 The measured EL intensity (open circles) and the corresponding least-square-fit (solid lines) of the OLEDs with different TPBi thicknesses at λ = 546 nm as a function of the rotating angle of the QWP with respect to polarizer.

PEM Method: Measurement of a Retardation of F8BT

For measuring an optical phase retardation, the phase modulation technique by a photoelastic modulator (PEM) has been used. Here, the PEM generates the time-dependent phase shift $A(t) = A_0\cos(\omega t)$ with an amplitude A_0 and a frequency ω. Suppose that B depicts the phase retardation of a sample at a certain rotation angle θ with respect to the optic axis of the PEM under crossed polarizers, as shown in Fig. S2, the transmitted intensity through the sample and the PEM is

$$E = \frac{1}{2}(1 \quad -1)\begin{pmatrix} exp\left[i\frac{B}{2}\right] & 0 \\ 0 & exp\left[-i\frac{B}{2}\right] \end{pmatrix}\begin{pmatrix} exp\left[i\frac{A_0}{2}\cos(\omega t)\right] & 0 \\ 0 & exp\left[-i\frac{A_0}{2}\cos(\omega t)\right] \end{pmatrix}\begin{pmatrix} 1 \\ 1 \end{pmatrix}$$

$$I = E^*E = \frac{1}{2}\{1 - J_0(A_0)\cos B\} + \frac{1}{2}J_1(A_0)\sin B\cos(\omega t) + \frac{1}{2}J_2(A_0)\cos B\cos(2\omega t) + \ldots$$

where $J_0(A_0)$, $J_1(A_0)$, and $J_2(A_0)$ denote the zeroth, first and second orders of the Bessel function, respectively. To eliminate the dependence of phase retardation B in the zeroth harmonics, we determine the modulation amplitude A_0 so that $J_0(A_0) = 0$. By using a lock-in amplifier, we can measure the intensities I_1 and I_2 of the first and the second harmonic terms, respectively.

$$I_1 = \frac{1}{2}J_1(A_0)\sin(B)$$

$$I_2 = \frac{1}{2} J_2(A_0) \cos(B)$$

Finally, we can determine the phase retardation B as rotating the sample (rotation angle θ) following as,

$$B(\theta) = \tan^{-1}\left(\frac{J_2 I_1}{J_1 I_2}\right)$$

As a result, for a given angle θ, we have measured the $B(\theta)$ as shown in Fig. 2(a). Now, to determine the phase retardation B_0 of the sample, which is a maximum $B(\theta)$ at θ = 0 (when the optic axis is parallel to that of the PEM), the transmitted intensity is modified as

$$E = \frac{1}{2}\begin{pmatrix}1 & -1\end{pmatrix}\begin{pmatrix}\cos\theta & -\sin\theta \\ \sin\theta & \cos\theta\end{pmatrix}\begin{pmatrix} exp\left[i\frac{B_0}{2}\right] & 0 \\ 0 & exp\left[-i\frac{B_0}{2}\right]\end{pmatrix}\begin{pmatrix}\cos\theta & \sin\theta \\ -\sin\theta & \cos\theta\end{pmatrix}\begin{pmatrix} exp\left[i\frac{A_0}{2}\cos(\omega t)\right] & 0 \\ 0 & exp\left[-i\frac{A_0}{2}\cos(\omega t)\right]\end{pmatrix}\begin{pmatrix}1 \\ 1\end{pmatrix}$$

$$I = E^* E = I_{dc} + J_1(A_0)\sin B_0 \cos(2\theta)\cos(\omega t) + \frac{1}{2}J_2(A_0)[1 - \cos(4\theta) + \{1 + \cos(4\theta)\}\cos B_0]\cos(2\omega t) + \ldots$$

Here, I_{dc} denotes the dc component of the transmitted intensity. For fitting the phase retardation B_0, we can used the following equation after comparing the above $B(\theta)$

$$B(\theta) = \tan^{-1}\left[\frac{2\sin B_0 \cos(2\theta)}{1 - \cos(4\theta) + \{1 + \cos(4\theta)\}\cos B_0}\right]$$

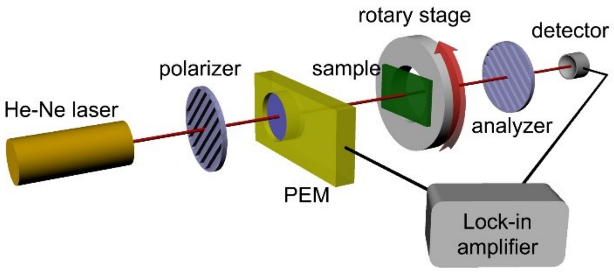

Fig. S2 Experimental setup for measuring the phase retardation of the sample.

Absorption Spectrum of F8BT and Emission Spectrum of TPBi

The absorption spectrum (solid line) of the pure F8BT film and the emission spectrum (dashed line) of the pure TPBi film were depicted in Fig. S3. Two spectra are strongly overlapped in the range from 410 nm to 500 nm, where the TPBi emitted light would be absorbed with polarization selectivity within the twisted F8BT layer.

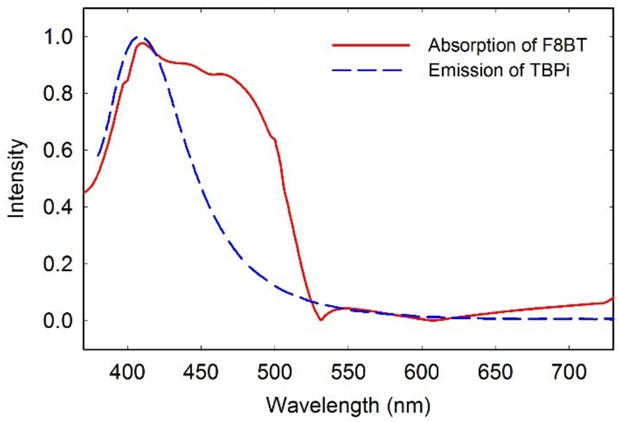

Fig. S3 Absorption spectrum (solid line) of the pure F8BT film and emission spectrum (dashed line) of the pure TPBi film.

Evaluation the *g* Factor

The general polarization state can be described by a combination of elliptical and random polarizations[2] and thus the EL polarization emitted from the F8BT or F8BT/TPBi layer is considered as a combination of such polarizations. In addition, the elliptically polarized light can be decomposed to orthogonal circularly polarized (CP) light with different amplitude: right-handed circular polarization (RHCP) and left-handed circular polarization (RHCP).[3] The sum of two CP components and the subtraction of them represent the longest length (l_a) and the shortest length (l_b) of the principal axes in the ellipse, respectively. For the evaluation of the *g* factor in the OLEDs with different TPBi thicknesses, light intensity at λ = 546 nm was measured as a rotating angle (θ) of QWP with respect to polarizer and presented in Fig. S4. For the analysis of the CPEL, we used the Jones matrix analysis for an elliptical polarization. For completely polarized light, the intensity as a function of the rotating angle is

$$I_{ideal}(\theta) = E^*E = \frac{1}{4}\left[\cos^2\phi\{3+\cos(4\theta)\} + 2\sin^2\phi\sin^2(2\theta) - \sin(2\phi)\{2\sin\delta\sin(2\theta) - \cos\delta\sin(4\theta)\}\right]$$

where

$$E = \begin{pmatrix}1 & 0\end{pmatrix}\begin{pmatrix}\cos\theta & -\sin\theta \\ \sin\theta & \cos\theta\end{pmatrix}\begin{pmatrix}e^{i\pi/4} & 0 \\ 0 & e^{-i\pi/4}\end{pmatrix}\begin{pmatrix}\cos\theta & \sin\theta \\ -\sin\theta & \cos\theta\end{pmatrix}\begin{pmatrix}\cos\phi \\ e^{i\delta}\sin\phi\end{pmatrix}$$

Considering the non-polarization, light intensity at λ = 546 nm was expressed by the following equation and the measured intensity was well fitted with the equation:

$$I^G(\theta) = \frac{I_{F,P}^G}{4}\left[\cos^2\phi\{3+\cos(4\theta)\} + 2\sin^2\phi\sin^2(2\theta) - \sin(2\phi)\{2\sin\delta\sin(2\theta) - \cos\delta\sin(4\theta)\}\right] + I_N^G$$

From the fitting parameters, we can evaluate two lengths l_a and l_b of the principal semi-axes of the elliptical polarization, and a rotating angle φ of the principal axes as expressed by the following equations:[4]

$$\tan(2\varphi) = \frac{2\cos\phi\sin\phi}{\cos^2\phi - \sin^2\phi}\cos\delta$$

$$l_a = \sqrt{\cos^2\phi\cos^2\varphi + \sin^2\phi\sin^2\varphi + 2\cos\phi\sin\phi\cos\delta\cos\varphi\sin\varphi}$$

$$l_b = \sqrt{\cos^2\phi\sin^2\varphi + \sin^2\phi\cos^2\varphi - 2\cos\phi\sin\phi\cos\delta\cos\varphi\sin\varphi}$$

Finally, the LHCP intensity (I_{LHCP}) and the RHCP intensity (I_{RHCP}) of the elliptical polarization were calculated, and the fitting values and the evaluated values were summarized in Table S1. Two *g* factors with/without considering the non-polarized emission (I_N^G) were evaluated. The measured *g* factor is in well agreement with the *g* factor with considering the non-polarized emission.

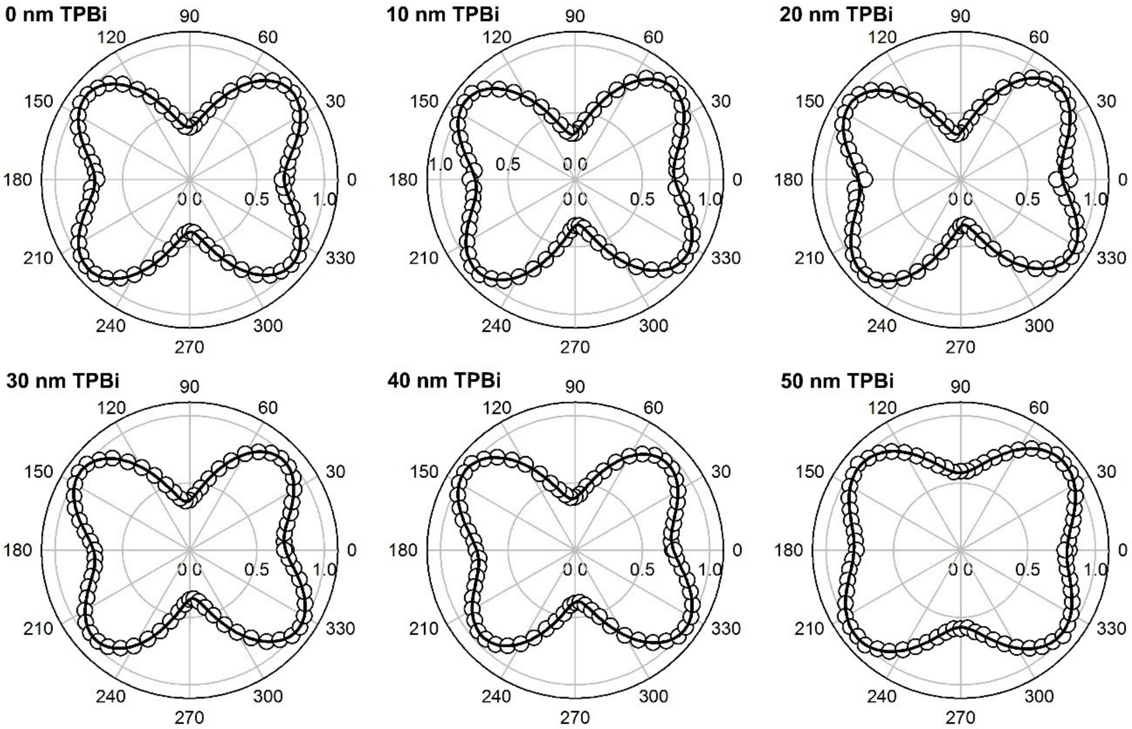

Fig. S4 The measured EL intensity (open circles) and the corresponding least-square-fit (solid lines) of the OLEDs with different TPBi thicknesses at λ = 546 nm as a function of the rotating angle of the QWP with respect to polarizer.

Table S1 Fitting parameters for an evaluation of the *g* factor in the OLEDs with different TPBi thicknesses under a rotation of QWP with respect to polarizer in Fig 4a.

TPBi thickness	0 nm	10 nm	20 nm	30 nm	40 nm	50 nm
$I^{G}_{F,P}$	0.927	0.966	0.954	0.928	0.878	0.635
ϕ (rad.)	0.179	0.212	0.224	0.213	0.227	0.228
δ (rad.)	-1.823	-1.519	-1.663	-1.116	-0.841	-2.248
I^{G}_{N}	0.082	0.052	0.066	0.072	0.127	0.370
I_{LHCP}	0.310	0.341	0.341	0.318	0.305	0.213
I_{RHCP}	0.153	0.142	0.135	0.146	0.134	0.104
$g_{EL}{}^{a}$	0.677	0.823	0.864	0.743	0.783	0.687
$g_{EL}{}^{b}$	0.575	0.744	0.760	0.643	0.608	0.318
Measured g_{EL}	0.582	0.736	0.751	0.633	0.597	0.298

[a] *g* factor without considering non-polarized contribution (I^{G}_{N}); [b] *g* factor with considering non-polarized contribution.

Müller Matrix Analysis for the *g* Factor

The polarization state of light propagating the birefringent medium is easily expressed by the Stokes parameters **S**, which is calculated by Müller matrix representing the birefringent medium. For calculation of the *g* factor, we assume that the F8BT layer is uniformly twisted in the film and divided to *N* sublayers.[4] Now, the twisted angle of the *i*-th sublayer depicts $\theta_i = \Phi z_i/d$, where Φ, z_i, and d are total twisted angle, distance from the hole transport layer, and the film thickness, respectively. The Müller matrix for the *i*-th sublayer with a phase retardation Γ is

$$\mathbf{M}(\Gamma,\theta_i)=\begin{pmatrix}1&0&0&0\\0&\cos(2\theta_i)&-\sin(2\theta_i)&0\\0&\sin(2\theta_i)&\cos(2\theta_i)&0\\0&0&0&1\end{pmatrix}\begin{pmatrix}1&0&0&0\\0&1&0&0\\0&0&\cos(\Gamma)&-\sin(\Gamma)\\0&0&\sin(\Gamma)&\cos(\Gamma)\end{pmatrix}\begin{pmatrix}1&0&0&0\\0&\cos(2\theta_i)&\sin(2\theta_i)&0\\0&-\sin(2\theta_i)&\cos(2\theta_i)&0\\0&0&0&1\end{pmatrix}$$

$$=\begin{pmatrix}1&0&0&0\\0&\cos^2(2\theta_i)+\sin^2(2\theta_i)\cos(\Gamma)&\cos(2\theta_i)\sin(2\theta_i)\{1-\cos(\Gamma)\}&\sin(2\theta_i)\sin(\Gamma)\\0&\cos(2\theta_i)\sin(2\theta_i)\{1-\cos(\Gamma)\}&\sin^2(2\theta_i)+\cos^2(2\theta_i)\cos(\Gamma)&-\cos(2\theta_i)\sin(\Gamma)\\0&-\sin(2\theta_i)\sin(\Gamma)&\cos(2\theta_i)\sin(\Gamma)&\cos(\Gamma)\end{pmatrix}$$

The light emitted at the *i*-th sublayer toward anode experiences the twisted birefringent medium.

$$S_{anode}=\prod_{j=i}^{N}M(\Gamma,\theta_j)S_e$$

Here, S_e and S_{anode} depict the Stokes vector of the emitted light at the *i*-th sublayer and that of the outgoing light to the anode. The light toward anode experiences reversely the twisted birefringent medium from the *i*-the sublayer to *0*-th sublayer and is reflected from the cathode. The reflected light experiences the whole twisted birefringent medium.

$$S_{cathode}=\prod_{j=0}^{N}M(\Gamma,\theta_j)\left[\prod_{j=0}^{i}M(\Gamma,\theta_j)\right]^{T}S_e$$

Here, $S_{cathode}$ depicts the Stokes vector of the outgoing light to the cathode. By the definition of the Stokes parameters, intensity I_L of the left-handed circular polarization is $(S_0-S_3)/2$ and the right-handed circular polarization is $(S_0+S_3)/2$. Also, using the definition of the *g* factor, $g=-2S_3/S_0$. We assume that the light emitted at a certain position within the emitting layer propagates toward anode or cathode with the same probability. Therefore, the final *g* factor was averaged over both propagating lights toward the anode and cathode. Using all measured parameters such as total twisted angle, total thickness, and birefringence, the *g* factor can be calculated as shown in Fig. 4d.

References

1 Y. Zhou, Z. He and S Sato, *Jpn. J. Appl. Phys.*, 1997, **36**, 2760–2764.
2 F. G. Smith and T. A. King, in *Optics and Photonics: An Introduction*, John Wiley & Sons, Ltd., New York, 2000, ch.6.
3 D. S. Kliger, J. W. Lewis and C. E. Randall, in *Polarized Light in Optics and Spectroscopy*, Academic Press, Inc., San Diego, 1990, ch.2.
4 D.-M. Lee, J.-W. Song, Y.-J. Lee, C.-J. Yu and J.-H. Kim, *Adv. Mater.*, 2017, **29**, 1700907.