Strongly Circularly Polarized Crystalline and β-Phase Emission from Poly(9,9-dioctylfluorene)-Based Deep-Blue Light-Emitting Diodes

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Dedicated to the memory of Professor Alasdair James Campbell

1. Introduction

In recent years, significant efforts have been made to improve the performance of organic light-emitting diodes (OLEDs), with a focus on both materials development[1–2] and device engineering.[3,4] Despite considerable advancements, the development of OLEDs with deep-blue (=420–450 nm) emission has presented an everlasting challenge.[5] Due to their excellent solution processability, wide bandgap, and high photoluminescence quantum efficiency, poly(9,9-dioctylfluorene) (PFO) and related polyfluorene homopolymers have been exploited as a major class of materials for deep-blue polymer light-emitting diodes (PLEDs).[6] PFO is an intensively characterized light-emitting polymer (LEP) known to exhibit several distinct solid-state polymorphs, including the glassy (amorphous) phase,[7–9] the β-phase,[9–11] crystalline α and α’-phases,[12,13] and the nematic liquid-crystalline phase.[7,14,15]

At the individual chain level, one of the key conformational differences between the PFO phases is the interunit torsion angle (ϕ). The subsequent interaction of polymer backbones with different ϕ and sidechain distributions results in distinct interchain spatial arrangements,[8,16] leading to the range of aforementioned polymorphs in the solid state. Such variations in the chain conformation and morphological phases directly affect the optical and electronic properties of the active layer including bandgap,[9,16,17] complex refractive index,[18,19] charge-transport properties,[9,15,20] emission wavelengths,[9,16,21] quantum yield,[22,23] and device stability under operational stress.[21] Glassy-phase PFO demonstrates a broad distribution of ϕ and stochastically varying di-octyl sidechain, with backbone conformational motifs in the glassy state[9] featuring ϕ ≈ 135°. Crystalline phase PFO can be formed by thermal annealing (TA), which promotes the formation of crystalline domains that exhibit a more ordered sidechain and backbone structure with ϕ ≈ 150°. From an OLED perspective, the formation of on-chain fluorenone defects due to electrical stress or photooxidation translates to a significant reduction in color purity and operational lifetime,[24,25] with such an impact particularly prone to glassy-phase PFO relative to structurally more ordered morphological phases. In contrast,
the β-phase adopts a well-defined zigzag planar conformation (φ = 180°).7,9 The spatially well-dispersed β-phase conformer with a narrower bandgap has been reported to act as a self-doping unit and therefore an exciton formation/harvesting site, which enables efficient energy transfer from nearby higher-energy glassy phase and crystalline phases for efficient light emission. The β-phase can be induced to the PFO bulk through judicious processing of post-deposition processing such as solvent vapor exposure (SVE),9 and has been shown to increase exciton coherence, diffusion length, and quantum efficiency, which ultimately improves OLED performance.21,28

An additional complication toward the commercialization of state-of-the-art deep-blue OLEDs is the light loss due to the incorporation of anti-glare filter in display applications.29,30 As demonstrated by prior works of our group and other groups,31,32 circularly polarized OLEDs (CP-OLEDs) can provide a simple yet effective solution. For CP-OLEDs that directly emit circularly polarized light with a luminance dissymmetry factor χ, the light loss factor γ is proportional to the rotatory strength, the chiral optical activity, and the dissymmetry of the molecular system. As shown in Figure S1, Supporting Information, for illustrative convention of handedness adopted in this work. Equation (1) reveals that for unpolarized emission (i.e., comprised of 50% left-handed and 50% right-handed light), the efficiency decreases by 50% when light propagates through the anti-glare filter (χ = 50%), whereas for fully CP emission (i.e., χ = 100%), no light loss occurs (χ = 0%). The magnitude of γ is proportional to the rotational strength, R, of the molecular system, which can be described by Equation (2):

\[ R_{ij} = \mu_{i,j} \cdot \mathbf{m}_{i,j} \quad \tag{2} \]

where i and j, respectively, stand for the initial and final states of an electronic transition of any individual chromophores, \( \mu \) and \( \mathbf{m} \), respectively, stand for the electric and magnetic transition dipole moments. In excitation coupled systems where electric dipole (\( \mu \)-\( \mathbf{m} \)) coupling between nearby chromophores dominates the chiroptical response, the rotational strength approximates (from a generic form as in Equation (2)) to Equation (3):

\[ R_{ij,2} = R_{ij} \cdot \mu_i \times \mu_j \quad \tag{3} \]

where \( \mathbf{r} \) represents the distance vector of both coupled transition dipoles. For simplicity, Equation (3) describes a dimerized case consisting of two interacting chromophores 1 and 2.13

An effective means to achieve circularly polarized luminance is to induce chiral phases into the LEP, either through the introduction of chiral sidechains or through blending with chiral small molecules.29,30,36 Impact of the morphology57 molecular weight55 and sidechain length35,38 on the chiroptical properties of polyfluorene (PF) derivatives have been extensively studied, although to the best of our knowledge there has only been one report on the associated device performance.38 By using poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) as an alignment layer, Geng et al reported a PF oligomer-based circularly polarized polymer light-emitting diode (CP-PLED) exhibiting a turn-on voltage \( V_{\text{on}} \) of 5 V, maximum current efficiency (CEmax) of 0.94 cd A⁻¹ and electroluminescence dissymmetry \( \gamma \) of 0.35. However, the handedness of \( \gamma \) was not easily controllable through the use of a particular sidechain enantiomer (R- or S-) which otherwise differ by length and branching motif.35 Specifically, oligofluorene with \(-3,7\text{-di}(\text{S}-2\text{-methylbutyl})\) sidechains exhibits a \( \gamma \) of +0.16, while oligofluorene with \(-3,7\text{-di}(\text{S}-3\text{-7-dimethyloctyl})\) exhibits an opposite handedness \( \gamma = -0.25 \). In contrast, it has been shown that \( \gamma \) of an achiral LEP polymer can be well-controlled by blending a chiral small-molecule additive following TA treatment to the active layer.29,30,36 CP-PLEDs with equal-and-opposite \( \gamma \) can be fabricated by blending enantiopure chiral small-molecule additives into the active layer, eliminating the synthetic demands of chiral sidechain polymers.29,30,36 It has previously been shown that chirality can be induced into β-phase PFO solution aggregates through control of the heating/cooling rate, solvent, and solution concentration.19,40 Despite these exciting observations, the induction of chiroptically active β-phase (hereafter “chiral β-phase”) domains into thin PFO films has not been previously reported, which shows central to the realization of high-performance CP-emitting PFO devices operational in the deep-blue spectral region. Here, we show that the chiral small molecule 1-aza[6]helicene (aza[6]H) can induce a chiral phase on crystalline PFO (circular dichroism, CD = -10000 mdeg) upon TA (T = 160 °C). Intriguingly, a subsequent SVE step promotes the formation of a chiral β-phase (CD = -8500 mdeg). Using such chiral β-phase-embedded PFO thin films as the active layers in PLEDs, we demonstrate the first deep-blue PLEDs exhibiting both strong CP emission (\( \gamma = -0.44 \)) and state-of-the-art device performance (CEmax = 1.23 cd A⁻¹). Compared to their semicrystalline-phase counterparts, embedding β-phase improved device efficiency, efficiency roll-off, and \( \gamma \) and may further provide mechanistic insight into the large chiroptical activity of polyfluorene materials.

### 2. Results and Discussion

First, we evaluate the impact of aza[6]H—a chiral small-molecule additive that we have previously demonstrated to induce a chiral phase in otherwise achiral polymers29,30,41—and TA on the UV–vis spectra of the PFO thin films (Figure 1). Compared to as-cast neat PFO, as-cast PFO:aza[6]H blend films demonstrate an additional high-energy absorption peak (λ = 261 nm), which corresponds to the absorption of aza[6]H. The use of TA above the polymer glass transition temperature to induce large chiroptical effects in PF (co-)polymer:aza[6]H blends has been evaluated by our group41,42 and we follow the optimized protocol in this work (160 °C for 5 min in a N₂ filled glovebox). It is important to note that as a TA step is essential to induce chiroptical activity, fully glassy-phase PFO:aza[6]H blends that demonstrate CP emission cannot be realized by our approach. As evidenced by Figure 1a, compared to the unannealed case, TA of the blend results in an apparent reduction in the intensity

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of the aza[6]H-related peak and a significant broadening of the main PFO $\pi$-$\pi^*$ absorption band ($\lambda = 385$ nm) toward both higher and lower energies. This indicates i) that the PFO content overall remains primarily a stochastically disordered conformation (i.e., with volume fraction of glass phase dominating the PFO bulk); and ii) that following TA the overall degree of chain aggregation and packing in PFO has been promoted (i.e., some isotropic PFO glass phase has transitioned to the crystalline phase). As shown in Figure 1b, as-cast PFO:aza[6]H blend thin films have negligible CD in the lowest-energy transition ($\lambda = 385$ nm) region. After TA (160 °C, 5 min; Figures S2 and S3, Supporting Information), a bisignate CD band ($\lambda = 406$ nm, $D \approx 11000$ mdeg) emerges, indicating strong excitonic coupling (see also further discussion below). Consistent with our previous studies, no obvious spectral changes are observed upon flipping or rotating the samples (Figure 1), strongly suggesting there are no contributions from either linear dichroism or linear birefringence.\[4,29\]

To induce $\beta$-phase segments within the chiral crystalline domains, the TA blend films were further treated with a toluene SVE process detailed by Shi et al.\[9\] As shown in Figure 2 and Figure S4, Supporting Information, SVE-treated films (SVE and TA+SVE) show a characteristic red-shifted absorption peak ($\lambda = 434$ nm) well resolved from the glassy and crystalline contributions. We find that the use of SVE alone (SVE only) results in thin films that are not chiroptically active (Figure S4, Supporting Information). Compared to the as-cast samples, both the TA and TA+SVE PFO:aza[6]H films exhibit strong chiroptical response (Figure 2). For the TA+SVE films, alongside the strong crystalline-phase CD peak at $\lambda = 402$ nm (CD $\approx -10000$ mdeg), a new CD peak emerges at $\lambda = 440$ nm (CD $\approx -8500$ mdeg), due to the induction of $\beta$-phase PFO segments. Similar to the crystalline-phase CD peak, the $\beta$-phase CD has negligible impact from linear dichroism–linear birefringence artefacts (Figure S5, Supporting Information). To disentangle the $\beta$-phase CD band from the overall chiroptical response we subtract the CD spectra obtained from a TA PFO:aza[6]H blend thin film before and after the SVE treatment. Interestingly, such a spectral subtraction exhibits a very large bisignate chiroptical response (Figure 2). We note that such an estimate serves only as an approximation: the SVE process converts a relatively small fraction of chiroptically active crystalline phase chain segments into $\beta$-phase, therefore, the total content of crystalline phase is not preserved between the two spectra.

Further to Equation (2), the dissymmetry factor associated with the transition dipole moments of an isolated chromophore can be expanded as below.\[42\]

$$ E = \frac{4R}{D} = \frac{4|\mu|\cos \theta}{|\mu|^2 + |m|^2} $$

where $D$ is the dipole strength and $\theta$ is the angle between $\mu$ and $m$.

Since the $\beta$-phase represents a highly planar PFO backbone conformation persisting over many repeat units (>15) with extended $\pi$-conjugation,\[7,17,23\] the intrinsic rotational strength from the single-chain $\beta$-phase backbone $R = |\mu|\cos \theta$ should be negligible, owing to both a weak $|\mu|$ and the orthogonal orientation of the transition dipole moments.\[43\] Instead, it seems the strong chiroptical activity of the chiral $\beta$-phase results from interchromophoric exciton coupling between adjacent planar polymer chains,\[44\] as presented in Equation (3). Through exciton coupling, the magnitude of the dissymmetry factor is no longer restricted by the relative magnitude of $\mu$ and $m$ of individual chromophores as in non-excitonically coupled systems.\[13,45–47\] Such enhanced rotatory strength in coupled systems originates from the coupled transition dipole moments of

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**Figure 1.** a) Normalized UV–vis spectra of as-cast neat PFO, as-cast PFO:aza[6]H, and thermally annealed PFO:aza[6]H films (all are ~120 nm on quartz substrates). b) CD spectra of as-cast blend films and TA blend films. The various measurement geometries in the figure legend are defined in Figure S1, Supporting Information. The inset shows the molecular structures of aza[6]H and PFO.
nearby chromophores with an appropriate mutual orientation, as described in Equation (3). We further note the split excited states, which occur as a result of exciton coupling, is compatible with our other experimental observations, including the broadened low-energy absorption band in Figure 1a and bisignate couplets in the CD spectra (Figure 1b; and the $\beta$-phase CD profile [marked as a green curve in Figure 2b] derived from spectral subtraction). We note that similar excitonically coupled polymers have been well-documented for, for example, poly(phenyleneethynylene)$^{[48-51]}$ and polythiophene$^{[52]}$ systems, featuring clear bisignate chiroptical response with asymmetric excition couplets.

The photoluminescence (PL) spectra of TA-only films of PFO:[M]-aza[6]H films show vibronic peaks at $\lambda_{0-0} = 433$ nm and $\lambda_{0-1} = 458$ nm (Figure S6, Supporting Information), which are typical of the crystalline phase.$^{[21]}$ Similar to the absorption spectra, the PL spectra of the TA+SVE films exhibit a red-shift relative to TA-only films ($\Delta \lambda_{0-0} = +5$ nm and $\Delta \lambda_{0-1} = +6$ nm), which indicates that the emission occurs from the more ordered and narrower bandgap $\beta$-phase contents (Figure S6, Supporting Information). Emission from the crystalline phase (TA-only films) is highly dissymmetric (Figure 3), with $g_{PL}$ $\approx +0.16$ at both 0–0 and 0–1 vibronic peaks. Interestingly, $\beta$-phase emission ($\lambda_{0-0} = 438$ nm) from the TA+SVE films has an even stronger dissymmetry ($g_{PL} = +0.23$). Thickness-dependent studies (Figures S7 and S8, Supporting Information) show an increase in thin film chiroptical activity, which plateaus at $\approx 200$ nm.

Non-resonant Raman scattering was performed to further validate the induction of $\beta$-phase as a result of the TA+SVE processing to the PFO:aza[6]H thin films. The spectra are shown in Figure S9, Supporting Information. Following the TA+SVE treatment, there is a clear increase in Raman intensity in the wavenumber region of $\approx 1100–1400$ cm$^{-1}$ and at $\approx 1581$ cm$^{-1}$. These observations indicate enhanced $\pi$-electron delocalization

Figure 2. a) Normalized UV–vis spectra of TA and TA+SVE PFO:[M]-aza[6]H; b) CD spectra of TA and TA+SVE PFO:[M]-aza[6]H: a vertical dashed line highlights the closely matched wavelengths at which $\beta$-phase absorption and CD features peak; $\Delta$CD presents a first-order approximation of the CD response of $\beta$-phase PFO by subtracting the TA-only from the TA+SVE spectra (see accompanying text for details). Film thickness $\approx 120$ nm.

Figure 3. CP-PL measurements of a) TA and b) TA+SVE PFO:[M]-aza[6]H films. $I_L$ and $I_R$ denote intensities of the left-handed and right-handed emission, respectively. Spectra are presented by peak-normalizing to the highest 0–0 transition intensity while retaining the relative intensity between $I_L$ and $I_R$. Films thickness $\approx 120$ nm.
due to the planarization of the conjugated backbone, and are consistent with previous reports on the conformational dependence of PFO Raman spectra.\cite{9,22} We note also from Figure S9, Supporting Information, that there are no noticeable differences between neat PFO and PFO:aza[6]H blend systems, which supports our conclusions that the aza[6]H does not disrupt the formation of \( \beta \)-phase.

We used noncontact-mode atomic force microscopy (AFM) to investigate the morphology of spin-coated PFO and PFO:aza[6]H thin films in their as-cast, TA-only, and TA+SVE states (see Figure S10, Supporting Information, and accompanying discussion). As evident from the AFM images, there exists no discernible aza[6]H phase segregation, irrespective of the processing conditions. In fact, the surfaces of all thin films considered are ultrasmooth, with root-mean-square roughness all under 0.5 nm. Such superior topographical uniformity (i.e., free from pin holes and apparent crystallites, etc.) affords the films as suitable candidates for OLED emissive layers.

As reported in our previous studies, aza[6]H (HOMO = −5.4 V) tends to aggregate toward the bottom of a film (i.e., the quartz or glass/ITO–polymer interface) during the TA step, where it can act as a hole trap in polyfluorene systems with deep-lying HOMO levels (e.g., PFO, HOMO = −5.8 eV).\cite{4} In the inverted OLED device architecture, which incorporates an air-stable electron-injection layer, hole injection occurs at the opposite interface to the one where aza[6]H accumulates. As a result, we have found devices with an inverted architecture to demonstrate significantly improved performance to the ones with a conventional architecture.\cite{4} CP-EL of PFO:aza[6]H-based devices was measured in an inverted structure of ITO/ZnO/PEIE/TPBi/PFO:aza[6]H/TCTA/MoO\(_x\)/Au (Figure 4). Chemical structures of all molecular materials used are shown in Figure S11, Supporting Information. We used 4,4′,4″-tris(carbazol-9-yl)triphenylamine (TCTA) as a hole-transport layer to shift the recombination zone further away from the aza[6]H accumulation region.\cite{4} Although polyethylenimine ethoxylated (PEIE) modified ZnO serves as an appropriate electron-injecting and hole-blocking layer in poly(9,9-diocetylfluorene-alt-benzothiadiazole (F8BT):aza[6]H devices,\cite{4} PFO:aza[6]H CP-PLEDs of the same device structure do not emit sufficient light to allow for an accurate determination of CP-EL. We attribute this to the shallow-lying LUMO level of PFO (LUMO = −2.6 eV), which results in a considerable injection barrier for electrons from PEIE modified ZnO (work function = 4.7 eV in the absence of interlayers, the current in PFO:aza[6]H CP-PLEDs is dominated by holes, and the recombination zone is located next to the PEIE/PFO interface. This results in a similar hole-trapping effect to that observed in conventional (i.e., non-inverted) CP-PLEDs, and also a low luminance. To improve device efficiency, we made use of an additional electron-transport layer,

Figure 4. a) Device structure and energy diagram showing all constituent layers of a PFO:aza[6]H CP-PLED. b) CP-EL spectra of crystalline (left) and \( \beta \)-phase-embedded (right) PFO:aza[6]H CP-PLED. \( I_L \) and \( I_R \) denote intensities of the left-handed and right-handed emission, respectively. Spectra are presented by peak-normalizing to the highest 0–0 transition intensity while retaining the relative intensity between \( I_L \) and \( I_R \). c) \( J–V–L \) and efficiency curves of crystalline (black) and \( \beta \)-phase-embedded (red) PFO:aza[6]H CP-PLED. Active layer thickness \( \approx 120 \) nm.
2,2’,2”-((1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi), and evaluated its impact using single-carrier devices (Figure S12, Supporting Information). Without the TPBi layer, the hole current density \( J_h \) is approximately two orders of magnitude higher than the electron current density \( J_e \) at a bias voltage of 5 V \( (J_h/J_e = 62:1) \) (Table S1, Supporting Information), which indicates that most holes flow through the device without recombining with electrons. Since TPBi exhibits a LUMO level (2.7 eV) that is midway between the ZnO/PEIE materials is not only important for further understanding this class of materials, but potentially as a design approach for other \( \pi \)-conjugated chiral organic materials. Indeed, by further building on prior studies, our data suggest exciton coupling as an important approach to realizing high performance materials for CP-OLEDs that retain both high optoelectronic performance and high chiroptical activity.

4. Experimental Section

4.1. azahelicene (aza[6]H) was prepared as previously reported and separated using preparative chiral HPLC.40 PFO \( (M_w = 59 \text{ kDa}) \) was provided by Cambridge Display Technology, Ltd. Solution Preparation and Thin Film Deposition: PFO was dissolved with a fixed ratio of aza[6]H (10 wt%) in toluene to form 20 mg mL\(^{-1}\) solution. Thickness of the thin films was controlled by spin speed (1500 rpm) and measured using a Dektak XT surface profiler. All samples were annealed at 160 °C for 5 min in nitrogen atmosphere (glovebox, \( \text{H}_2\text{O} < 0.1 \text{ ppm}, \text{O}_2 < 0.1 \text{ ppm})

**Organic Light-Emitting Diode Fabrication and Characterization:** Prepatterned ITO substrates were sequentially rinsed in an ultrasonic bath with acetone, isopropyl alcohol (IPA) (both from Sigma Aldrich) and Hellmanex III (Hellma GmbH), and deionized water before deposition of top layers. The ZnO was deposited on the cleaned ITO films was carried out using a Park NX-10 AFM in noncontact mode with a moderately rigid cantilever (PPP-NCHR, force constant of 41 N m\(^{-1}\))

**5. Conclusion**

The rich variety of accessible solid-state phases of PFO allow for determination of fundamental relationships between chain conformation and photophysical/chiroptical signatures, as well as access to blue-emitting CP-PLEDs. Here, we present the first deep-blue \( \beta \)-phase PFO CP-PLED with both high performance (CE = 1.23 cd A\(^{-1}\)) and high dissymmetry \( \left| \langle g_{EL} \rangle \right| = 0.44 \) where the chiroptically active \( \beta \)-phase domain is formed by sequential treatment of TA and SPE. Especially given the planarized polymer backbone, we attribute the large chiroptical activity of this “chiral \( \beta \)-phase” to exciton coupling between adjacent polymer chains. The central role that exciton coupling appears to play in the large chiroptical response for PFO-based materials is not only important for further understanding this class of materials, but potentially as a design approach for other \( \pi \)-conjugated chiral organic materials. Indeed, by further building on prior studies, our data suggest exciton coupling as an important approach to realizing high performance materials for CP-OLEDs that retain both high optoelectronic performance and high chiroptical activity.

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**Table 1. Summary of device performance of PFO:aza[6]H CP-PLEDs.**

| Device variants                    | \( V_{on} \) [V at 1 cd A\(^{-1}\)] | CE [cd A\(^{-1}\)] | PE [lm W\(^{-1}\)] | \( L_{max} \) [cd m\(^{-2}\)] |
|------------------------------------|--------------------------------------|--------------------|---------------------|-----------------------------|
|                                    |                                       | Maximum            | At 1000 cd m\(^{-2}\) | Maximum                    |
| Semicrystalline                    | 4.0                                   | 1.13               | 0.97                | 0.81                        |
|                                    |                                       | At 1000 cd m\(^{-2}\) |                     | 0.51                        |
| Further \( \beta \)-phase embedded| 4.4                                   | 1.23               | 1.20                | 0.61                        |
|                                    |                                       | At 1000 cd m\(^{-2}\) |                     | 0.60                        |
|                                    |                                       |                     | 1.38                | 1583                        |

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prior to any data acquisition. During the measurement, samples were kept in a N₂ chamber to avoid degradation.

Circular Dichroism, Circularly Polarized Electroluminescence, and Circularly Polarized Photoluminescence: The CD data were collected using a Chirascan 100 (Applied Photophysics) instrument. Left-handed and right-handed CP emission spectra from the blend thin films were collected using a linear polarizer and a zero-order quarter-wave plate (λ = 405 nm) prior to the detector of an FLS-1000 fluorospectrometer. Both the excitation beam and the detector were at 45° (collected using a linear polarizer and a zero-order quarter-wave plate, respectively). A similar method was used to analyze the CP-EL spectra. EL spectra from the PLED were recorded using an Ocean Optics USB 2000 charge-coupled spectrophotometer. All CP-EL measurements were carried out after measuring with only a linear polarizer to ensure negligible linear polarization or random polarization was present which may otherwise impact the accuracy of circular polarization measurement.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare the following competing financial interest(s): A.J.C. and M.J.F. are inventors on a patent concerning chiral blend materials (WO2014016611).

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
β-phase, circular polarization, interchromophoric exciton coupling, inverted organic light-emitting diodes, organic light-emitting diodes, polyfluorene

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