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## **Electronic Supporting Information**

**Controlling Condensed State Structures of Different Polar Conjugated Polymer Polyfluorenes (PFs) by Applying External Electric Field across Solution with Polar Solvent THF** 

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1. Molecular Weight and Polydispersity Index of Samples.

Figure S1. The distribution of retention time of different polar conjugated polymers' solutions. a) PFO; b) PPFOH; c) PODPF.

Table S1. M	<b>Iolecular</b> W	eight and l	Polydispersity	Index of	the Samples

samples	PFO	PPFOH	PODPF
molecular weight	70460	14700	65903
(Mw) (g mol-1)			
polydispersity index	1.17	3.17	1.66
(PDI)			

### 2. Experiment Measurement Methods.

2.1. Photoluminescence (PL) Spectra. Photoluminescence spectra were taken with a

fluorescence spectro-photometer (Shimadzu RF-5301PC) equipped with a xenon lamp

as excitation source. The measurements were recorded in a range from 400 nm to 600 nm with excitation wavelength at 384 nm at room temperature.

**2.2. Light Scattering Measurements.** An ALV/CGS-3 (made in Germany) light scattering spectrometer equipped with an ALV/LSE-7004 multiple- $\tau$  digital correlator was used in the Light Scattering (LS) measurement. The JDS-Uniphase solid-state He-Ne laser with the output power of ca. 22 mW at the operating wavelength of 632.8 nm was used as the light source. The LS cell was held in a thermostat filled with purified and dust-free toluene.

The aggregation structure with polymer nonlinear property can be assessed by means of SLS. In the measurement, the average scattered light intensity  $I_{(q)}$  could be described as a function of the scattering wave vector  $q=4\pi\lambda sin(\theta/2)$ , where  $\theta$  is scattering angle and  $\lambda$  is the wavelength, we set the accessible scattering angles range from 50° to 150°, so the range of q was in 12.6 µm<sup>-1</sup><q <28.7 µm<sup>-1</sup>. The collected light intensity  $I_{(q)}$  depends mainly on the particle form factor  $P_{(q)}$  and the aggregate structure factor  $S_{(q)}$ , they could connect followed by the formula:

$$I_{(q)} = K P_{(q)} S_{(q)}$$
(1)

Here,  $P_{(q)}$  contains information on the optical properties of the individual particle, whereas  $S_{(q)}$  quantifies mass correlations within the aggregation, K depends on the optical properties of the light scattering device. In stable systems without any spatial correlation between the monomer positions,  $S_{(q)} \approx 1$ . In this case,  $P_{(q)}$  can be determined easily from direct measurements of the scattered light intensity, since  $I_{(q)} \propto P_{(q)}$ . For an aggregated system, however, the structure factor  $S_{(q)}$  is directly related to the fractal dimension of the clusters formation,  $d_f$  quantifies how compact the fractal aggregates are, meanwhile, q was in the range of  $R_{agg} > q^{-1} > R_g$ , where  $R_{agg}$  and  $R_g$  are the root of radius of gyration for the aggregations and single chain, respectively.

$$I_{(q)} \propto S_{(q)} \propto q^{-df}$$
 (2)

The normalized scattering intensity autocorrelation function  $g^2(t)$  was measured in DLS, where *t* is the decay time.  $g^2(t)$  is related to the normalized first-order electric field time correlation function  $g^1(t)$ , and they could establish the Siegert relation as  $g^2(t) = 1 + \beta |g^1(t)|^2$ .  $g^1(t)$  is the Laplace transformation of the decay time distribution function A(t) of the decay rate *t*, and the A(t) can be required using the CONTIN analysis.

**2.3. Raman Spectra Measurement.** Raman spectra with JY-T64000 Raman spectrometer, grating type equipped with2017, argon ion gas laser was used to explore the conformation change, its excitation wavelength was at 514.5 nm.

**2.4. X-ray Diffraction (XRD) Measurement.** XRD analysis was performed using a high-resolution X-ray diffractometer (SmartLab, Rigaku Corporation, Japan) with monochromatized Cu K $\alpha$  radiation ( $\lambda = 0.15406$  nm).

**2.5. Transmission Electron Microscope (TEM) Measurement.** Transmission electron microscope (TEM) and high-resolution transmission electron microscope (HR-TEM) images were obtained by JEM-2100 (made in Japan) to explore the chain aggregation structure. The magnified lattice fringes represented the domain of ordered aggregation.

#### 3. Raman Spectra at High Wave Number Region of PFO Films.

The high wavenumber region (1000–1650 cm<sup>-1</sup>) shown in Figure S2 corresponds to the main chain motion of PFO chain. Especially, the peak at around 1604 cm–1 softly shift to short wavenumber under the external electric field strength 10KV/cm, which proved PFO chain transitioned towards more planar chain geometry with the longer effective longer conjugated length as shown in Figure S2b.



Figure S2. Raman spectra at high wave number region of PFO films. a) 1000-1800cm<sup>-</sup> <sup>1</sup>; b) 1550-165cm<sup>-1</sup>. The films were prepared from the 5mg/mL PFO/THF precursor solution subjected the action external electric field.

# 4. PL Spectra of PODPF High Concentrated Solutions under the Action of External Electric Field.

It was found that PODPF chain conformation in high concentrated 5 mg/mL solution didn't change at all even under applying strong strength of external electric field.



Figure S3. PL spectra of PODPF high concentrated 5mg/mL solutions under external electric field at 10KV/cm.