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Supplementary Note 1. Strategy for highly dissymmetric chiral active layer

A diverse set of optoelectronic conjugated organic semiconductors, namely, poly(3-hexylthiophene-2,5-diyl) (P3HT), poly(9,9-dioctylfluorene-alt-bithiophene) (F8T2), poly[2,6-(4,4-bis(2-ethylhexyl)-4H-cyclopenta-[2,1-b:3,4-b']dithiophene-alt-4,7-bis(thiophen-2-yl)benzo-2,1,3-thiadiazole] (PCPDTTBTT), poly{2,2'-[(2,5-bis(2-octyldodecyl)-3,6-dioxo-2,3,5,6-tetrahydropyrrolo[3,4-c]pyrrole-1,4-diyl)dithiophene-5,5'-diyl-alt-thieno[3,2-b]thiophen-2,5-diyl} (PDBT-co-TT), poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene vinylene] (MEH-PPV), and poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b;4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene-)-2-carboxylate-2,6-diyl] (PTB7-Th) were blended with chiral additives, namely, 1,1'-Binaphthyl-2,2'-diamine ((*R*)- or (*S*)-BN), (13bR)-5,6-dihydro-5-(trans-4-propylcyclohexyl)-4H-dinaphtho[2,1-f:1',2'-h][1,5]dioxonin (R5011 or S5011), (2,2-dimethyl-1,3-dioxolane-4,5-diyl)bis(diphenylmethanol) ((*R*, *R*)- or (*S*, *S*)-TADDOL), (*R*)- or (*S*)-2-octyl 4-[4-(hexyloxy)benzoyloxy]benzoate (R811 or S811), 2-amino-3-mercaptopropionic acid (*L*- or *D*-cysteine), and 4'-[(*S*)-2-methylbutyl]biphenyl-4-carbonitrile (CB15), which were previously used as chiral active layers^{S1-6} was tested in the onset of this study. The molecular structures of the various conjugated polymers and chiral additives used to fabricate the homochiral active layers are provided in **Supplementary Figs. 1 and 2**. All solutions were spin-coated on quartz plates for CD and UV-vis spectroscopic analyses; the results of the as-cast and thermally annealed films are compared in **Supplementary Figs. 3-8**. The fluorene-based conjugated polymer exhibited a strong CD activity when the chiral additives were blended in. Unanticipated CD bands were observed in the films of PCPDTTBTT. The CPDT-based polymer displayed a strong and distinct Cotton effect when doped with (*S*)-BN, CB15, R811, (*S*, *S*)-TADDOL, and S5011. Chain rearrangement by thermal treatment at temperatures above the mesophase transition temperature induced significant chiroptical activities in both

conjugated polymer-based thin films. The films with strong CD bands showed a large Cotton effect in the absorption region of the solid-state conjugated polymeric films, regardless of the chiral dopants. This behavior is indicative of chirality transfer from chiral small molecule additives to entire active layer systems. The CPDT-based polymer doped with R5011 or S5011 exhibited unusual chiroptical properties, presumably because of the large helical twisting powers of R5011 and S5011, as well as efficient intermolecular interactions between the polymer and additives^{S6}. The impressive chirality transfer and amplification in π - π^* transitions suggest high-performance broadband applications with strong electric dipole-allowed transitions (i.e., large absorption coefficient).

In contrast, negligible changes in CD spectra were observed with films of other conjugated polymers. Several semiconductor-additive combinations exhibited weak Cotton effect (i.e., P3HT with (*S*)-BN and R5011, MEH-PPV with (*S*)-BN and S5011, PDBT-co-TT with (*S*)-BN, CB15 and S5011, and PTB7-Th with (*S*)-BN, CB15 and S5011), but their chiroptical activities were very weak (< 100 mdeg) when compared with the aforementioned strong CD-effective films. Moreover, the CD-effective range was localized to the high energy molecular electronic transition of chiral small molecule additives; it disappeared after thermal treatment at temperatures above the glass transition temperature (T_g) of the semiconductors because of chiral additive sublimation. This localization was caused by chiroptical effects that originated from intramolecular magneto-electric dipole coupling or weak intermolecular excitonic coupling in only partially twisted stacked chiral additive molecules; it indicated that the chiral additives could not induce π -conjugated semiconducting chromophores to contain any helical or twisted (i.e., chiral) arrangement for chiral excitonic coupling in major π - π^* transitions.

The chirality transfer in polymer films with strong CD activity is related to their molecular and supramolecular ordering because fluorene-based conjugated polymers are well-known liquid crystalline π -conjugated molecules^{S7,8}. We investigated differences in molecular packing

or orientation phenomena in the solid state (**Supplementary Fig. 9**). The as-cast polymer films manifested featureless structures because of weak and short-range molecular ordering. Pronounced birefringent textures were observed in the annealed films of F8T2 and PCPDTTBTT. Both films exhibited distinct birefringent textures that were significantly affected by chain rearrangement during the thermal treatment, leading to highly crystalline and long-range molecular ordering. This result strongly indicated that the significant chiroptical activities caused by chiral additives emerged with supramolecular and long-range molecular ordering beyond simple π - π interactions.

Both conjugated polymers exhibited very intense CD bands in their respective absorption ranges. However, PCPDTTBTT displayed broadband UV-vis and CD spectra with a more red-shifted absorption edge at ~ 800 nm. Fluorene-based polymers are frequently used in luminescence devices, rather than photon-absorbing devices, because of their high quantum yield and fast radiative recombination^{S6,9-11}; these properties contrast with the CPDT-based conjugated polymers used in solar cells and photodetector devices^{S12-14}. Therefore, we fabricated the intrinsic chiral active layer using the CPDT-based conjugated polymer for broadband circular polarization (CP) detection and explored the corresponding extraordinary chiroptical activities and spin angular momentum (SAM) detection device physics, with the intention to use them in real-time spin imaging prototype devices.

Supplementary Note 2. Circular dichroism study

The addition of chiral dopant R5011 or S5011 and subsequent thermal annealing resulted in extremely strong chiroptical properties (**Fig. 2a**, **Extended Data Fig. 2**). The doped films exhibited mirror-image CD signals according to the chirality of the dopant with large CD intensity ($> 20^\circ$) and dissymmetry factor (g) (~ 0.8) in thin films (160 nm). The supramolecular chirality of the main absorption band of the polymers originated from efficient chirality inspirations within highly crystalline mesoscopic alignments. **Supplementary Fig. 10b, c** shows that the removal of dopants after thermal annealing was clearly discernible in the UV-vis spectra, which demonstrated that the chiral dopants acted as a template for helical stacks of polymer chains during chain rearrangements, rather than forming complexes with the polymer chains or crystal itself.

We observed the CD-silent feature in as-made (i.e., before annealing) doped films and annealed doped films at a higher temperature (290°C), compared with the clearing temperature of the polymer (**Supplementary Fig. 10d, e**). Notably, the CD signals were saturated as the dopant content reached 50 wt% (**Extended Data Fig. 3a**). Additional introduction of chiral additives could not affect further changes in chiroptical effects, which indicated that the superposed chiral dopants did not cause a sudden reduction of chiroptical effects. **Extended Data Fig. 3b** shows the thickness-dependent chiroptical effects, which were quantitatively estimated as g values by normalizing the CD to the unpolarized absorbance of the sample. The extraordinarily high g values of *ca.* 0.8–1.2 exhibited by the doped polymeric films in their CD spectra were independent of film thickness (> 100 nm). Notably, the thin films showed small g values (~ 0.1) in very thin films (< 60 nm), but these values remained high with respect to other conjugated polymers except the fluorene series. This behavior may be related to the strong surface effect on oxide substrates, leading to the formation of different morphologies during solidification and chain rearrangement processes.

Supplementary Note 3. Thermal property study

Thermogravimetric analysis and differential scanning calorimetry (DSC) were used to characterize the phase behavior and crystallization properties of the conjugated polymers as a function of temperature (**Supplementary Fig. 12**). PCPDTTBTT lost 5 wt% as the temperature increased to 430°C and decomposed by initial loss of alkyl substituents, which suggested high thermal stability. The peaks and valleys in DSC curves correspond to the release or intake of thermal energy that occurs during phase transitions. The DSC thermogram of the polymer PCPDTTBTT revealed the T_g at ~140°C and the onset of cold crystallization at 189°C. Two peaks observed between 220°C and 255°C indicated that the polymer contained two distinct mesophase states. The first peak at ~235°C corresponded to the melting of the solid to mesophase and the second peak corresponded to the transition between mesophases. Finally, the onset of the mesophase–isotropic transition appeared at 257°C, when the polymer became a disordered, liquid phase. Therefore, the optimized annealing temperature of 250°C used in this study allowed the polymer to attain a long-range ordered supramolecular structure, thereby enabling intense chiroptical properties with the addition of chiral dopants.

Polarized optical microscopy (POM) studies with crossed linear polarizers revealed that the thin films annealed at a temperature above the clearing temperature did not have any locally anisotropic and birefringent domains (**Supplementary Fig. 13**). Similarly, the as-cast films showed no birefringence because they lacked long-range ordering of the polymer chains. In contrast, clear Schlieren textures were observed after thermal treatment for the mesophase transition. The annealed chiral doped films displayed Schlieren textures almost identical to the textures of neat films, regardless of the ratio of chiral dopants (**Supplementary Fig. 14**). This indicated the absence of highly ordered cholesteric stacking, such as the stacking observed in

double-spiraled textures (with a certain helical pitch) of chiral nematic films formed on rubbed substrates or sandwiched liquid-crystal cells^{S15}.

Supplementary Note 4. Microscale morphology analysis

Scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used to observe the surface morphological structure of the chiral active layer at the microscale (**Fig. 2b, c, Supplementary Figs. 15 and 16**). Before thermal annealing, undoped films and films with high chiral doping content showed morphologies similar to small nanobeads. After thermal annealing, the aggregates rearranged into a fibril-like organization with a rough surface (root-mean-square height: 1.9 nm) in the absence of chiral dopants. The long, one-dimensional structures at the submicron scale were responsible for the substantial optical anisotropic properties observed in POM images. In contrast, the presence of chiral small molecules during thermal annealing resulted in the rearrangement of constituent bead-shaped aggregates into smooth and homogeneous, wavy arrays of string-like features (root-mean-square = 0.7 nm), which differed from the amorphous and rugged aggregates of the neat films. The surface comprised nanofibrils with mean width of *ca.* 10–20 nm and length < 1 μm . Thus, the introduction of chiral small molecules resulted in the rearrangement of “worms” of constituent beads into wavy fiber arrays with smooth surfaces. The nanofibrils were arranged parallel to the surface, which may have been influenced by the nature of the free space^{S1}. Transmission electron microscopy (TEM) measurements revealed flattened and oriented surfaces in a wavy rod-like fibril array organization with a heterogeneous chain rearrangement (**Supplementary Fig. 17**).

Intermediate morphological features were observed in species annealed below the mesophase transition temperature (**Supplementary Fig. 18**). SEM images of doped films annealed at 180°C exhibited much shorter fibrillar structures with remaining nanobeads, which indicated that the nanofibrils had formed by the aggregation of existing nanobeads. Indeed, the intermediate structures without submicron one-dimensional structures showed no enhancement of chiroptical properties, regardless of chiral additive doping (**Supplementary Fig. 10e**). The

magnified view of doped films annealed at 250°C in high-resolution AFM images indicated that the nanofibrils were composed of a string of beads *ca.* 4–5 nm in diameter (**Supplementary Fig. 19**). In addition, several nanofibrils with width of *ca.* 5–10 nm aggregated into bundled nanofibers.

Cross-sectional SEM images were observed to investigate the effects of interfacial interactions among the substrate, films, and free space on the morphology of active layers across the film depth (**Fig. 2d**, **Supplementary Figs. 20 and 21**). Molecular rearrangements (ranging from bead-like morphologies to rod-like features) were observed throughout neat polymer films after thermal annealing, resulting in very rough surfaces and cross-sections. In contrast, chiral small additives resulted in much smoother surfaces and cross-sections in the annealed state. Notably, the nanofibrils preferred vertical ordering on the substrate, but tended to arrange in parallel at the interface with air, where the arrangements were more distinct in doped films. Similar erratic features were observed in large-scale and side-view cross-sectional SEM images in the presence of both enantiomeric dopants (**Supplementary Fig. 22**).

Supplementary Note 5. X-Ray diffraction study

Sub-microscale molecular packing was investigated using X-ray diffraction techniques. Powder X-ray diffraction measurements revealed structural changes in neat and doped polymer films before and after thermal treatments (**Supplementary Fig. 23**). The annealed films showed a sharp peak at $2\theta = 7.30^\circ$ for out-of-direction molecular stacking ($d = 12 \text{ \AA}$), which corresponded to interdigitated packing of linear alkyl side chains or chain backbone repeat distance, rather than π - π stacking of conjugated planes. The diffraction peaks caused by chain rearrangement during thermal annealing were clearer and more distinct in neat films than in doped films, which indicated reduced crystallinity or a decrease in existing molecular ordering through the construction of another crystalline order after introduction of chiral small molecules. The 3D packing structures and geometries of polymer chains were further elucidated by 2D GIWAXD studies. Before thermal annealing, both neat and doped polymer films exhibited a polycrystalline nature with several well-defined ring patterns without any chain ordering preference in both in-plane and out-of-plane directions (**Supplementary Fig. 24**). **Figure 2e** shows that the 2D GIWAXD patterns of the annealed neat films had highly oriented X-ray reflections with the strongest (001) diffraction peak at $q_{xy} \approx 0.52 \text{ \AA}^{-1}$ in the out-of-plane direction ($d = 12 \text{ \AA}$, similar to the diffraction pattern observed in powder X-ray diffraction), which corresponded to the typical interchain distance of conjugated D-A polymers, such as diketopyrrolopyrrole (DPP)-, isoindigo (ISO)-, and rylene diimide-based polymers. The polymer chains crystallized in the triclinic *P1* space group with unit cell lattice constant lengths a , b , and c of 12.0, 9.5, and 12.7 $\text{\AA}$ and lattice constant angles α , β , and γ of 97° , 95° , and 90° , respectively. When advancing the interpretation that chiroptical activities, film morphologies, and molecular packing are related to a surface effect, questions arise regarding molecular arrangements and crystallinities according to film thickness direction. **Supplementary Fig. 25** shows 2D GIWAXD patterns as a function of incident angle α_i . The

neat polymer films exhibited even distribution of crystallinity according to film depth, which indicated a weak surface effect of molecular packing during the chain rearrangement.

Chiral small molecule doping caused the emergence of distinctive crystalline changes; a new diffraction pattern was observed in relation to typical columnar structures with a fade-out of the triclinic polymeric crystals (**Fig. 2f**, **Supplementary Fig. 26**). Because the dopant-only films displayed an amorphous ring-pattern halo in the thin-film state (**Supplementary Fig. 27**), we suspect that the new diffraction pattern resulted from chain rearrangement of the CPDT-based polymer in the presence of the chiral small molecules. The patterns were not differential according to chiral composition. However, we concluded that the patterns were distinct from the “usual” packing, namely, conventional lamella stacking of conjugated polymers; they exhibited unique supramolecular long-range ordering. Analysis of the GIWAXD results revealed that the small-angle region reflection corresponded to a tetragonal 2D columnar phase with helical π -stacked assemblies (**Fig. 2g**, **Supplementary Fig. 28**). The center-to-center distance between helices (corresponding to the helix outer diameter), calculated from the position of the (100) Bragg reflection, was 45 Å. The intrahelix π -stacking distance estimated from the position of (010) reflection was 3.83 Å. Therefore, the polymer chains with chiral small molecule additives and thermal treatment presumably did not form the regular layer-like structures exhibited by common conjugated polymers; instead, they formed supramolecular helical columnar structures. The 2D GIWAXD patterns as a function of incident angle revealed the crystallinities of the in-plane helical ordering according to film depth (**Supplementary Fig. 29**). These patterns showed weak diffraction peaks at a beam incident angle of 0.13°, which indicated a strong surface anchoring effect on in-plane helical ordering during the chiral templating process.

Supplementary Note 6. XPS, FTIR, and ToF-SIMS studies

We performed X-ray photoelectron spectroscopy (XPS) and Fourier-transform infrared spectroscopy (FTIR) on the film surface to systematically investigate the helical rearrangement mechanism and dynamics of CPDT-based polymer films according to chiral templating. Dopant introduction and the annealing process did not significantly alter the S 2p and N 1s spectra because these peaks resulted from the polymer (**Supplementary Fig. 30**). The O 1s peak observed for the neat polymer thin film was likely caused by the oxidation of branched alkyl chains during measurement, which was conducted under ambient conditions; thermal annealing reduced the elemental oxygen content. In contrast, when chiral dopant was introduced into the polymer films, the O 1s spectrum exhibited a clear C–O–C peak at 532.5 eV, which originated from the molecular structure of the dopant. However, after thermal annealing, the peak almost disappeared; the peak caused by C–O–H increased in intensity (**Extended Data Fig. 4a**). This result implied that the chiral dopant caused chiral templating for supramolecular helical ordering. Most molecules were removed by sublimation at high temperature, as indicated by the sharply reduced overall atomic concentrations of carbon and oxygen after thermal annealing (**Extended Data Table 1**). Similarly, the C–O peak of the C 1s spectrum of the doped polymer film was noticeably less intense after thermal annealing, which implied removal of the chiral dopant.

The chiral templating was also confirmed in the FTIR spectra of the blended thin film, where distinct peaks derived from the chiral dopant disappeared after thermal annealing (**Supplementary Fig. 31**). The peaks at $\sim 2,900\text{ cm}^{-1}$ in the polymeric films originated from the 2-ethylhexyl side chains of PCPDTTBTT, while the peaks at $\sim 1,500\text{ cm}^{-1}$ originated from ring stretching vibrations of the thiophene, cyclopentadiene, and benzene groups. The peaks from chiral dopant almost disappeared after thermal annealing. However, the peak at $1,080\text{ cm}^{-1}$

¹ corresponding to the C–O stretching vibration remained visible after thermal annealing; it originated from the few remaining oxygen-containing moieties involved in interactions between the polymer chains and chiral dopants.

ToF-SIMS under negative polarity helped us to establish the diffusion-dependent effect of the chiral templating as a function of film depth (**Supplementary Fig. 32**). The introduction of chiral dopant strongly enhanced oxygen-containing peaks (O^- , OH^- , and C_2OH^-). In the as-cast mixtures, the oxygen-containing peaks and chain fragments ($\text{C}_3\text{H}_5\text{S}^-$, C_2HS^- , CHNS^- , and C_4HS^-) directly attributed to the polymer chain did not change according to depth, which indicated that polymer and chiral dopant were homogeneously distributed throughout the thin film without any phase separation in dopant or polymer blends after solvent removal. In addition, the doped films exhibited increased signals of S-containing chain fragments with an additional peak corresponding to CSO^- . The appearance of this peak may be related to ring structure degradation into R–S, R–SN, and R–SO moieties because of strong noncovalent bonding between the S/N content of CPDT/BT/thiophene in the polymer and the O content of the chiral dopants^{S16,17}. The typical oxygen peaks almost disappeared after thermal annealing, which was consistent with the surface XPS observation of decreased C–O–C peak intensity. Notably, unlike the O-containing peaks, the S-containing peaks greatly changed according to depth even after dopant sublimation, which indicated that the chiral templating phenomenon was more dominant at the free surface because of the surface anchoring effect and difficult dopant diffusion near the substrate (**Extended Data Fig. 4b**). From the aforementioned results, we concluded that the chiral dopants induced long-range intrachain helical ordering in supramolecular nanofabric structures via strong intermolecular interactions, such as $\text{S}\cdots\text{O}$ and $\text{N}\cdots\text{O}$, during polymer chain rearrangement.

Supplementary Note 7. Effect of device structure on dissymmetric properties

Unlike optical properties, high-performance SAM detection requires electronic sensors with a delicate device configuration design. The g_{ph} values and specific detectivity (D^*) as functions of the chiral additive concentration and film thickness were investigated to determine the impact of the chiral templating phenomenon on chiroptical properties (**Extended Data Fig. 5**). Chiroptical activities increased as the dopant concentration increased, with saturated g_{ph} values in equivalent weight blends; this finding indicated that extensive dopant loading would be necessary to maximize the chiral templating effect via noncovalent bonding with the polymer backbone. Notably, the strongly chiral insulators did not affect photocurrent conversion performance because of efficient sublimation in the thermally activated chiral templating process. Therefore, we decided to incorporate 50 wt% dopant to maximize the spin detection capabilities without reducing electrical readout performances.

However, spin-to-photocurrent conversion performances were substantially affected by the active layer thickness. The D^* decreased with increasing active layer thickness because thick active layers limited charge carrier mobility and led to the recombination of photogenerated charge carriers before they reached the relevant electrodes^{S18,19}. The g_{ph} values increased with increasing active layer thickness; they reached a maximum at ~400 nm but were not saturated, in contrast to the dissymmetry factors in the absorption spectra. The observed mismatch of optimal thickness in CD and CP photoresponsivity (**Extended Data Fig. 3b, 5b**) presumably occurred because the incident photon-to-photocurrent conversion process was strongly dependent on photodetector device configuration.

Spin-electronic signal conversion properties were investigated for different device configurations to better understand the chiroptical activities and the photocurrent conversion principle. Modulation of the effective photoconversion area and fast carrier transport without recombination is critical for a high dissymmetry factor in SAM detection. Photodetectors with

a symmetric gold electrode configuration displayed weak parabolically shaped current–voltage (I – V) curves that resulted from the two small Schottky barriers connecting the symmetric electrodes (**Supplementary Fig. 34**). Strong dissymmetry in the photocurrent g factors occurred as a function of applied voltage; this finding was not observed under normal light or dark current. The effective absorption area for the photocurrent contribution was a narrow depletion region at each electrode–semiconductor interface, which was caused by exciton formation with high binding energy, limited carrier mobility, and fast recombination before collection. Under the reverse electric field, the effective photocurrent conversion area was localized on the top electrode; this resulted in higher selectivity toward spin direction because of the enhanced chiroptical activities on the free surface produced by the strong surface anchoring effect.

Asymmetric structures were studied to better understand control of the depletion region (**Supplementary Fig. 35**). They exhibited rectifying diode behavior with a rectification ratio of 10^2 , which resulted in leakage-current suppression and photocurrent enhancement. The dissymmetry factors were significantly increased in the reverse bias region (**Fig. 3e**). Under a high negative bias condition, the dissymmetric behavior dramatically diminished, which originated from excessively wide interfacial barriers under high reverse bias and resulted in unclear photocurrent because the dark current was a more dominant component of the current flow^{S20}.

Zinc oxide layers (that are efficient hole blockers) sharply increased the rectification ratio ($> 10^3$) (**Supplementary Fig. 36**) confirming the proposed mechanism of optoelectronic transduction. The dark current decreased by approximately one order of magnitude, leading to better specific detectivities. Furthermore, the enhancement of rectifying properties led to increased g values throughout the bias region, especially in the high negative biased condition. The increased energy barrier suppressed charge injection under the strong bias, while

maintaining the energy cascade to facilitate extraction of photogenerated carriers. In addition, chiroptical properties were enhanced in a self-powered condition because no external bias was needed to promote exciton dissociation due to sufficient built-in potential.

A fullerene derivative was introduced into the chiral active layer for interfacial trap-assisted hole tunneling injection because the chiral templating for the formation of helical structures decreased with increasing addition of fullerene derivative (**Supplementary Fig. 37**). This behavior was attributed to the absence of long-range ordering of polymer chains caused by the presence of the immiscible solid-state fullerene within the polymeric phase, which prohibited the formation of helical ordering through interactions with chiral dopants^{S21}. To intensify photomultiplication, a small amount (1 wt%) of phenyl-C71-butyric acid methyl ester (PC₇₀BM) was blended in the optimized devices. The trapped electrons induced interfacial band bending, which resulted in efficient hole tunneling injection from the external circuit under light illumination; this led to a photomultiplication (**Supplementary Fig. 38**)^{S22}. The optimized structures displayed excellent photodetection properties without decreasing the g value ($|g_{ph}|$ value remained at 1.2 and D^* over 10^{11}) throughout the negative voltage sweep region (**Supplementary Fig. 39**). Importantly, dramatic enhancement of dissymmetric photodetection was observed under the forward bias condition, rather than under the reverse bias condition. Distinct interfacial band bending according to the spin direction of incident photons occurred at the chiral active layer–MoO₃/Au interface because of the surface anchoring effect; this led to increased hole tunneling injection from the anode source and subsequent photocurrent conversion. Thus, the chiroptical activities could be modulated by systematic design of the photodetector configurations, thereby broadening the highly dissymmetric region for SAM detection in both photoconductive mode and photovoltaic photodiode operations (**Extended Data Fig. 6**).

Supplementary Note 8. Degree of circular polarization

The Stokes vector is a four-element real vector that describes polarized or partially polarized light. The four Stokes vector elements are expressed as follows:

$$S = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} E_{0,x}^2 + E_{0,y}^2 \\ E_{0,x}^2 - E_{0,y}^2 \\ 2E_{0,x}E_{0,y} \cos \delta \\ 2E_{0,x}E_{0,y} \sin \delta \end{bmatrix},$$

where $E_{0,x}$ and $E_{0,y}$ are the respective amplitudes of the x and y fields, and δ is the phase difference between the two orthogonal components of the light wave^{S23}. The S_0 parameter describes the total intensity, S_1 describes the amount of light polarized in the vertical or horizontal direction, S_2 describes the amount of light polarized at $\pm 45^\circ$ to the x/y -axis along the direction of propagation, and S_3 describes the circular polarization properties of the light wave.

The DoCP is the fraction of the intensity attributable to circularly polarized light states, with 0 corresponding to no circular polarization and 1 corresponding to complete circular polarization. Mathematically, DoCP is defined using the following Stokes vector elements:

$$\text{DoCP} = \frac{S_3}{S_0} = \frac{2E_{0,x}E_{0,y}}{E_{0,x}^2 + E_{0,y}^2} \sin \delta.$$

Therefore, when measuring the DoCP of light while rotating the retarder angle, the DoCP values according to the phase difference appear as a sine wave. In our high-performance CP detectors, the sine trend that appeared in the recorded photocurrent upon rotation of the retarder angle was in good agreement with the results estimated from the sine equation.

The overall photocurrent based on both circular and linear polarizations as a function of the rotation angle of the retarder was described by the following expression:

$$I_{PC} = C \sin(2\theta + \varphi_c) + L \sin(4\theta + \varphi_l) + D$$

where θ is the rotation angle of the retarder, C is the amplitude of the circularly polarized light, φ_c is the phase of the circularly polarized light, L is the amplitude of the linearly polarized light, φ_l is the phase of the linearly polarized light, and D is a polarization-independent constant background response^{S24}. The recorded photocurrents were accurately fitted with the first periodic term, which represents the helicity-resolved component of the incident beam. The second periodic term related to linear polarization was negligible.

Supplementary Note 9. Chiroptical properties in various optical set-ups

To confirm that the excellent chiroptical properties originated from true optical activity, rather than structural chirality, we examined CD behavior while changing the light–matter interaction condition (i.e., azimuthal sample rotation, sample flipping, and incidence angle of the exposed beam) (**Fig. 4b**, **Extended Data Fig. 7**). According to electronic CD interpretation based on Mueller matrix analysis, the CD signal of thin film could be specified as follows^{S25}.

$$CD_{obs} \approx CD_{iso} + \frac{1}{2} (LD' \cdot LB - LD \cdot LB') + (-LD \cos 2\theta + LD' \sin 2\theta) \sin \alpha \\ + (P_x^2 - P_y^2) \sin 2\alpha (-LB \cos 2\theta + LB' \sin 2\theta)$$

where CD_{iso} is the isotropic CD; LD (LD') and LB (LB') are the linear dichroism and linear birefringence associated with the x and y (x' and y' ; with a $+45^\circ$ shift of x and y axes) axes, respectively. θ is angle of sample rotation around the optical axis, α is associated with the residual birefringence of photoelastic modulator of CD machine, and a is the azimuth angle of the photomultiplier with respect to the vertical axis. P_x^2 and P_y^2 are related to the transmittance of the detector in x and y axes.

To record isotropic electronic CD of the solid sample, CD_{iso} , we observed the CD spectra of chiral polymeric films by flipping the sample by 180° with respect to the vertical axis to remove the CD based on the macroscopic anisotropic effect^{S26}. The second periodic term is related to the interference between linear dichroism and linear birefringence and the sign could be inverted by the sample flipping. We confirmed that there were almost negligible differences among CD spectra throughout the spectral range, indicating that the chiroptical properties originated from the intrinsic chirality of intrachain helical stacks, rather than from linear dichroism and birefringence^{S27,28}. In addition, we recorded CD spectra by rotating the azimuthal angle of the mounted sample around the optical axis of the incident light. The third and fourth periodic terms are related to the residual static birefringence of the PEM, α , and the

characteristics of the photomultiplier as a planar polarizer, respectively^{S25}. The sample rotation along the optical axis (θ) corrected the CD_{iso} measurement by removing the artificial CD by technical issues of the spectropolarimeter.

We also investigated linear polarization effects on the chiroptical activity of the electronic CP detectors by illuminating linearly polarized light as a function of rotating azimuthal angle of the polarization axis of the linear polarizer (**Supplementary Fig. 53**). Constant photocurrent signals obtained regardless of the spatial azimuthal angle showed no correlation of our chiral films with linear dichroism, which is expected from thin cholesteric orders of fluorene-based polymer chains^{S29}. In addition, the electrical readouts of both circularly and elliptically polarized light were tested under horizontal-axis variations of CP light (**Fig. 4d**). The CP detectors showed highly sensitive DoCP detection in any azimuthal orientation between the irradiated devices and polarization axis.

Supplementary Note 10. Mueller matrix spectroscopic ellipsometry

In the Mueller–Stokes description, the light–sample interaction is described by a linear relationship between the Stokes vectors of incident (S_i) and outgoing (S_o) light through a 4×4 Mueller matrix (M) that implies the following sample characteristics:

$$\begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix}_{out} = \begin{bmatrix} M_{00} & M_{01} & M_{02} & M_{03} \\ M_{10} & M_{11} & M_{12} & M_{13} \\ M_{20} & M_{21} & M_{22} & M_{23} \\ M_{30} & M_{31} & M_{32} & M_{33} \end{bmatrix} \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix}_{in}$$

To analyze the optical properties, the Mueller matrix must be reduced to its basic six optical properties. This is accomplished via matrix logarithm:

$$m = -\ln(M)$$

$$m = \begin{bmatrix} \alpha & \beta & \gamma & \xi \\ \beta & \alpha & \mu & \nu \\ \gamma & -\mu & \alpha & \eta \\ \xi & -\nu & -\eta & \alpha \end{bmatrix}$$

where α is the isotropic absorption; β , γ , and ξ are the linear dichroism at $0^\circ/90^\circ$, linear dichroism at $45^\circ/135^\circ$, and circular dichroism, respectively; and η , ν , and μ are the linear birefringence at $0^\circ/90^\circ$, linear birefringence at $45^\circ/135^\circ$, and circular birefringence, respectively^{S30}.

We mainly investigated the Mueller matrix elements of m_{03}/m_{30} to quantify CD of the films comparing it for transmission and reflection modes. In the transmission mode, the CD spectra inferred from m_{03} and m_{30} exhibited signals similar to the signals observed in conventional CD measurements. It should also be noted that the spectra exhibited uniform intensity regardless of light incident angle and sample azimuthal rotation. Conversely, in the reflection mode, the CD components were almost negligible at all oblique incidences.

Supplementary Note 11. Chiral media interaction with incident light at oblique angles

Similar to the effects of linear dichroism and birefringence on chiroptical activities, the incident angle of the polarized beam can be a dominant factor in SAM detection. In some inhomogeneous chiral media, optical responses do not change when exposed to a circularly polarized beam under azimuthal rotation. However, there are fundamental differences in terms of how chiral media react with obliquely incident electromagnetic waves between homogenous matter with true optical activity and an inhomogeneous system with a periodic helicoidal structure. In typical cholesteric-like structures, the CD intensity increases with decreasing incident angle because the maximum circular reflection always occurs at normal incidence when the wave vector of light is parallel to the helical axis. In addition, the spectral optical response shifts towards shorter wavelengths as the incident angle increases according to the expression $\lambda = \lambda_0 \cos\theta$, where θ is the angle of incidence and λ_0 is the peak position at normal incidence^{S31}. Thus, the operating range and DoCP sensitivity of the sensor devices were critically limited under oblique incident conditions of both circularly and partially polarized light^{S32}.

Supplementary Note 12. Selective CP detection in array

A 30×30 array of photodiodes was illuminated with a CP beam in “2/3” or “S/ε” patterns while alternately changing the handedness of the CP beam (i.e., right-handed circular polarized [RCP] beam to left-handed circular polarized [LCP] beam). CP beams with opposite polarization helicities were alternately irradiated at the oblique incident angle of 10° (**Fig. 5a–c** and **Extended Data Fig. 9a**) and normal incident angle (**Extended Data Fig. 9b–d**) while the photocurrent was recorded. Mode 1 corresponded to irradiation with the LCP beam to form the “S/ε” pattern, while the “2/3” pattern was formed by irradiation with the RCP beam. Similarly, Mode 2 corresponded to irradiation with the RCP beam to form the “S/ε” pattern and irradiation with the LCP beam to form the “2/3” pattern. Accordingly, M1, and M2 were defined as follows:

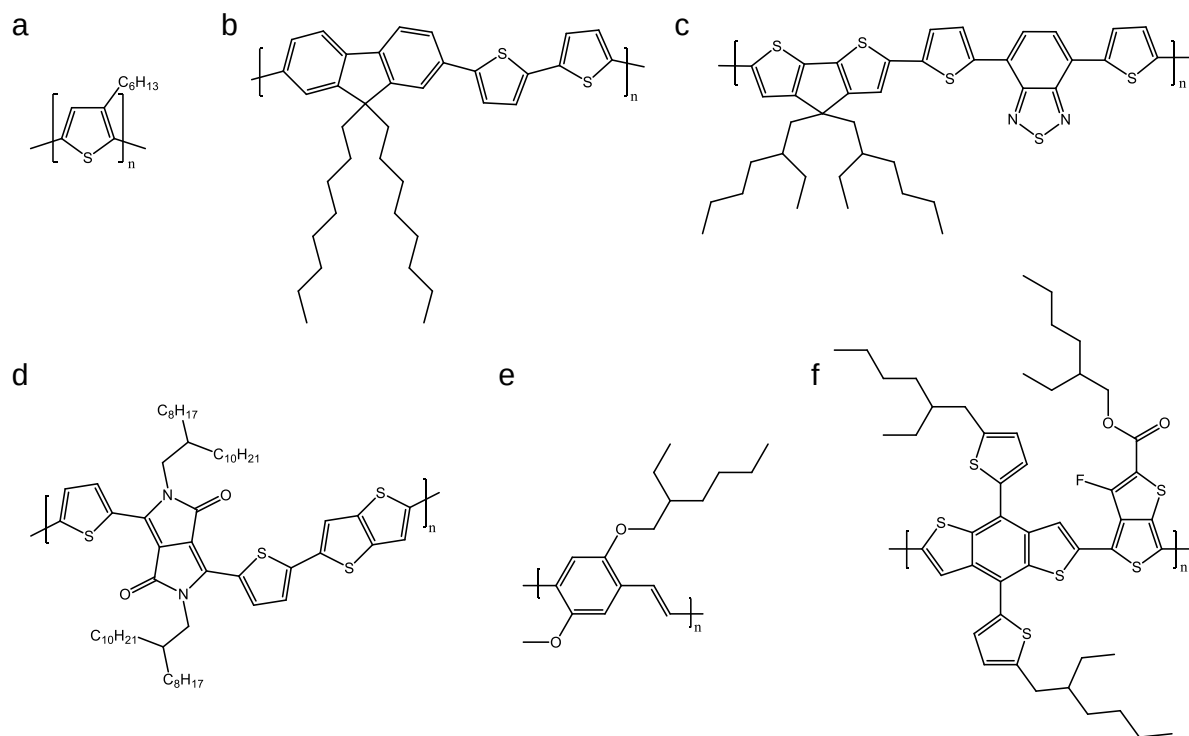
$$M1 = \frac{I_{light}}{I_{dark}} \text{ at Mode 1}$$
$$M2 = \frac{I_{light}}{I_{dark}} \text{ at Mode 2}$$

In the CP detection mode, $M1 - M2$ became negative with the “S/ε” pattern and positive with the “2/3” pattern because RCP generated more photocurrent in the array photodiodes. Therefore, patterns were clearly distinguished and observed during imaging.

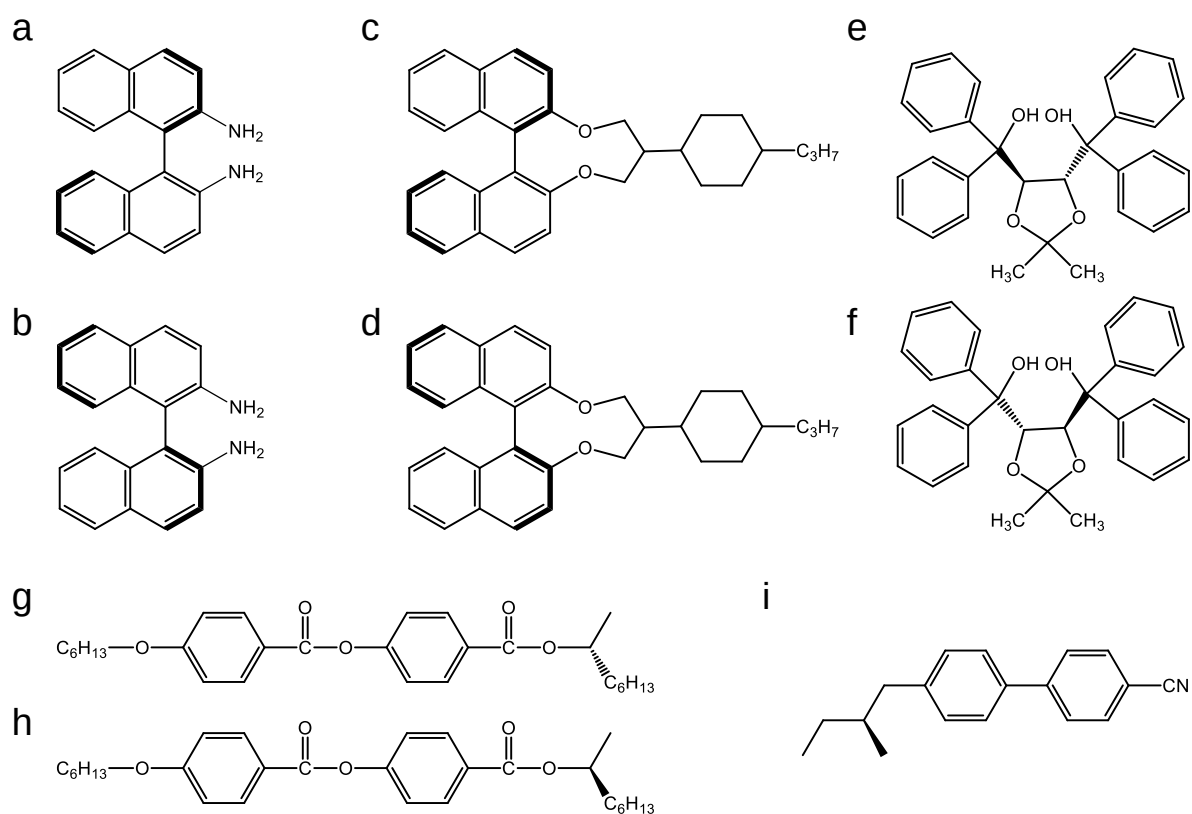
However, in the conventional photodetection mode, assuming that the photodetectors could not distinguish LCP and RCP light, the mean photocurrent value, $\frac{M1+M2}{2}$, was plotted and imaged. In this case, the “2/3” or “S/ε” pattern could not be distinguished and an image resembling a “heart/four-leafed clover” appeared. This result is a simple example of the ability to transmit hidden information using CP light; it may have future uses in cryptography.

Supplementary Note 13. Passive matrix CP real-time detection

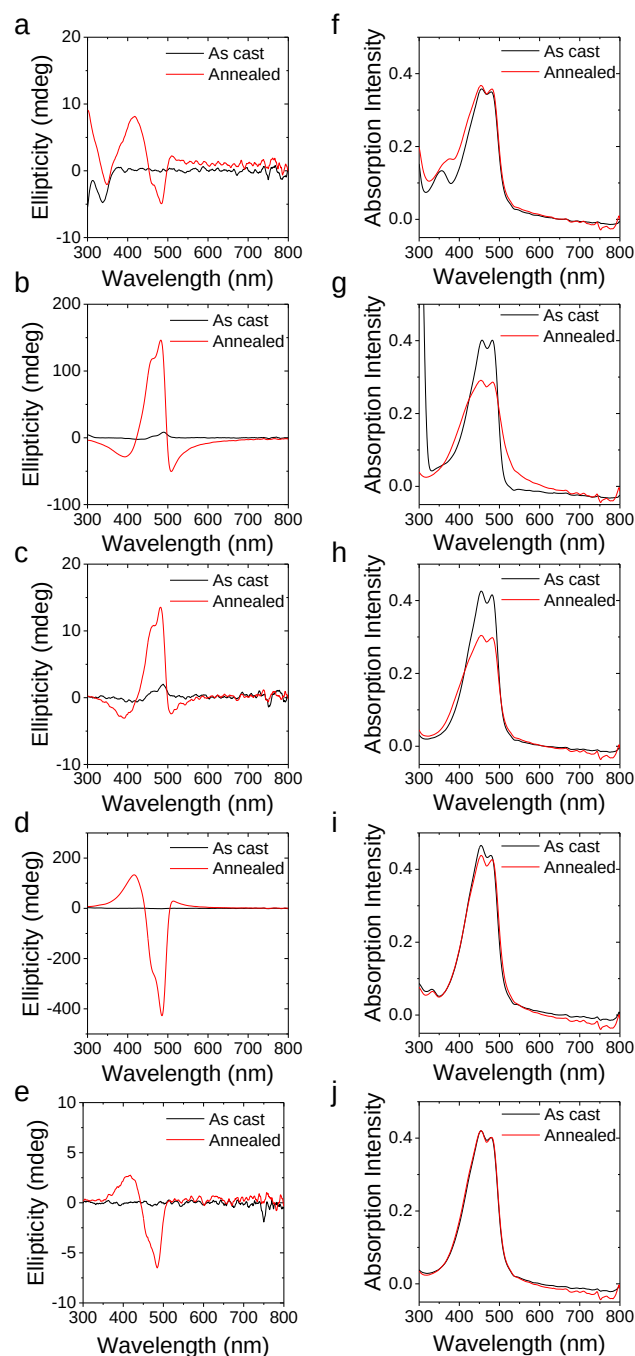
Our devices, which consisted of two lined, gridded electrodes with a blocking layer, transporting layer, and optimized chiral media, were driven by the passive matrix driving method using 5 V of input voltage for multiple signal detection demonstration using multiplexing in the grid structures. The ADC values were determined by the resistance of our sensor device and external reference resistors in the signal addressing units, according to the following relationship: $ADC_{ij} = R_{ext}/(R_{ext} + R_{ij}) \cdot A$, where ADC_{ij} corresponds to each pixel device of the matrix sensor, R_{ext} is the resistance of the external resistors, R_{ij} is the resistance of each pixel device, and A is the signal addressing constant. When a pixel was illuminated by a CP beam, the resistance of the chiral active layers decreased compared with the reference resistors, leading to enhancement of terminal conversion values. Optimization of the resistance and dissymmetry factors of the chiral media provided a three-state electrical readout, which was demonstrated in ternary quantum application experiments (final digital conversion values: 0 for dark state, 1 for LCP beam, and 2 for RCP beam).



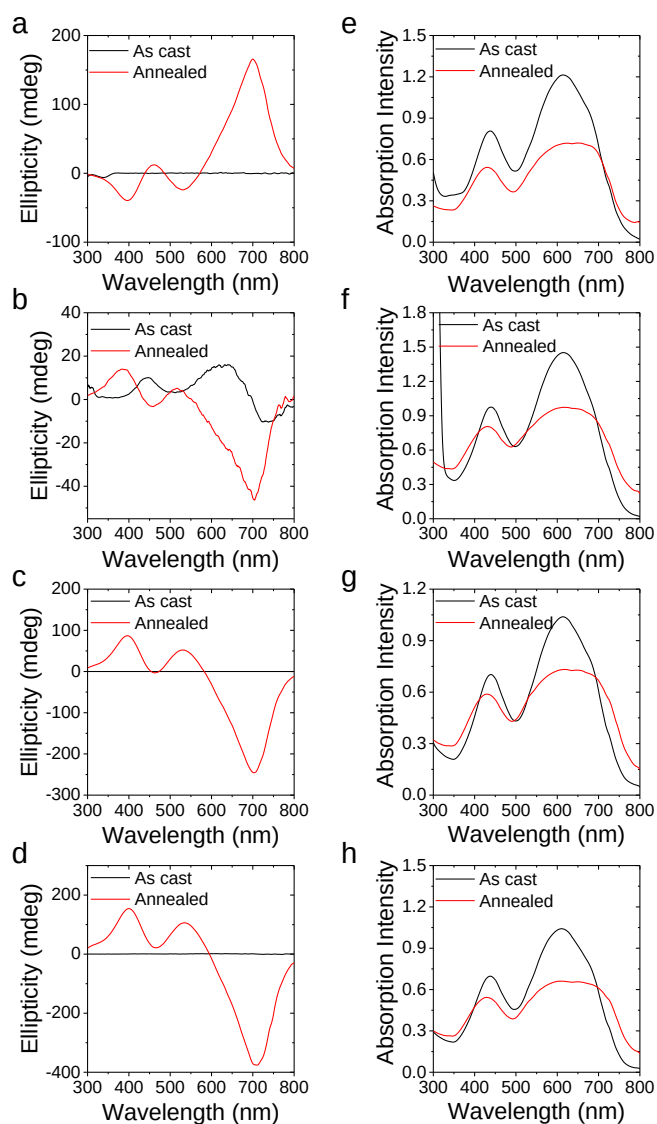
Supplementary Fig. 1 | Molecular structures of the tested conjugated polymers. a–f, Molecular structures of P3HT (a), F8T2 (b), PCPDTTBTT (c), PDBT-co-TT (d), MEH-PPV (e), and PTB7-Th (f).



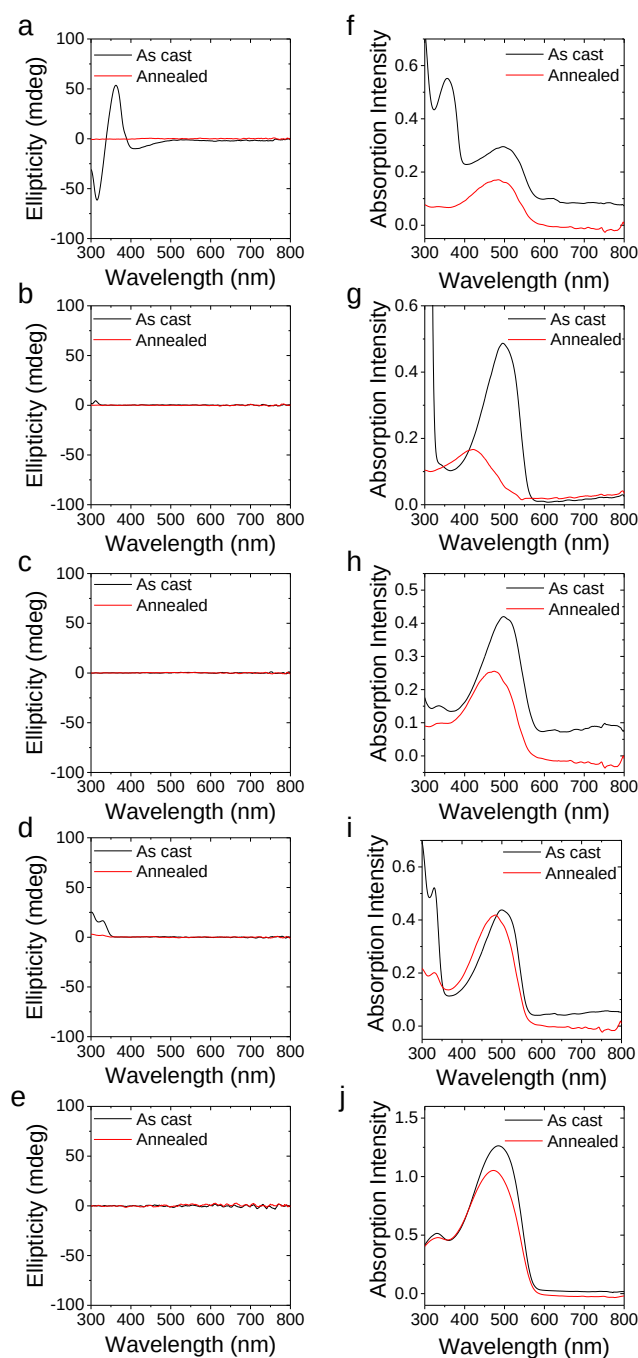
Supplementary Fig. 2 | Molecular structures of various chiral additives. a–k, (R)-BN (a), (S)-BN (b), R5011 (c), S5011 (d), (R, R)-TADDOL (e), (S, S)-TADDOL (f), R811 (g), S811 (h), and CB15 (i).



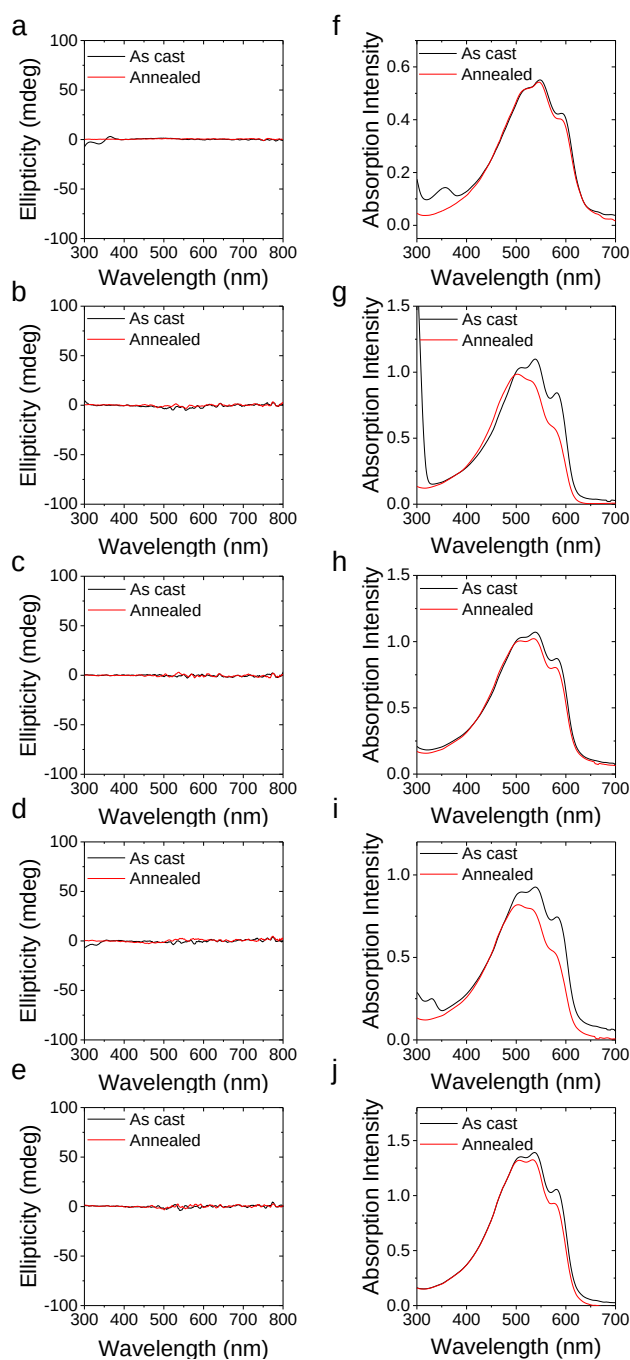
Supplementary Fig. 3 | CD and UV-vis absorption spectra of F8T2 with various chiral dopants. a–e, CD spectra of (S)-BN (a), CB15 (b), R811 (c), S5011 (d), and (S, S)-TADDOL (e) doped F8T2 thin films before and after thermal annealing. **f–j**, UV-vis absorption spectra of (S)-BN (f), CB15 (g), R811 (h), S5011 (i), and (S, S)-TADDOL (j) doped F8T2 thin films before and after thermal annealing.



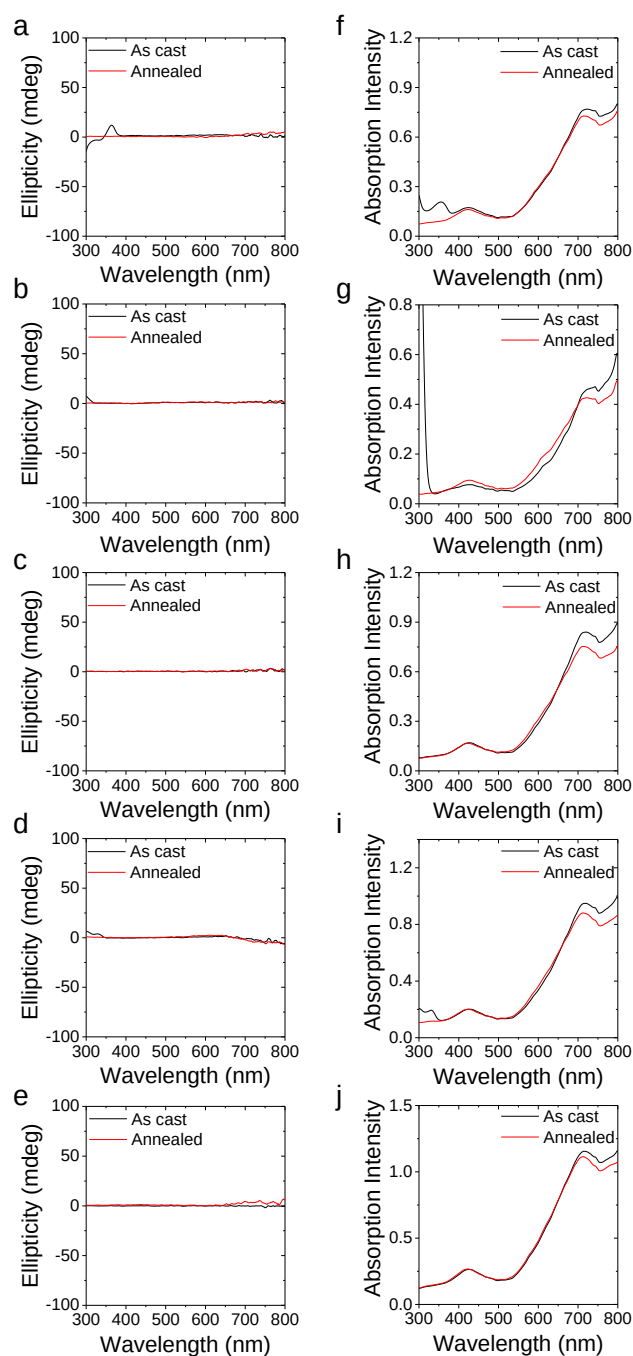
Supplementary Fig. 4 | CD and UV-vis absorption spectra of PCPDTTBTT with various chiral dopants. a–d, CD spectra of (S)-BN (a), CB15 (b), R811 (c), and (S, S)-TADDOL (d) doped PCPDTTBTT thin films before and after thermal annealing. **e–h**, UV-vis absorption spectra of (S)-BN (e), CB15 (f), R811 (g), and (S, S)-TADDOL (h) doped PCPDTTBTT thin films before and after thermal annealing.



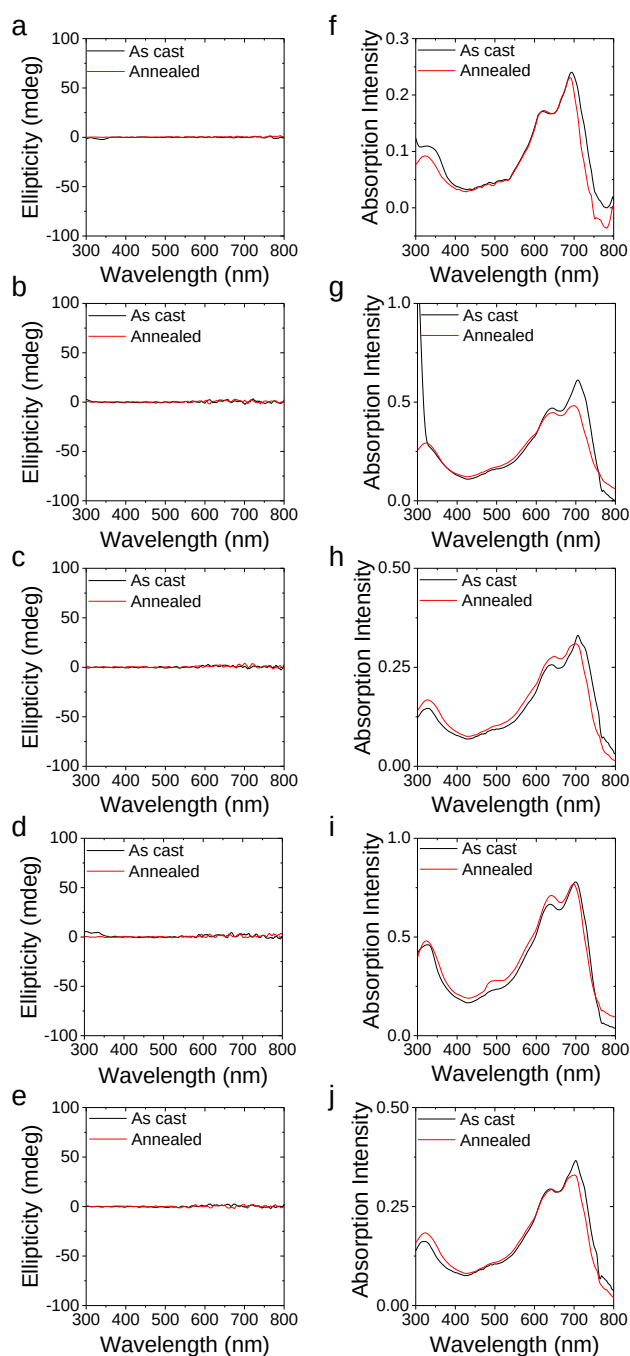
Supplementary Fig. 5 | CD and UV-vis absorption spectra of MEH-PPV with various chiral dopants. a–e, CD spectra of (S)-BN (a), CB15 (b), R811 (c), S5011 (d), and (R, R)-TADDOL (e) doped MEH-PPV thin films before and after thermal annealing. **f–j**, UV-vis absorption spectra of (S)-BN (f), CB15 (g), R811 (h), S5011 (i), and (R, R)-TADDOL (j) doped MEH-PPV thin films before and after thermal annealing.



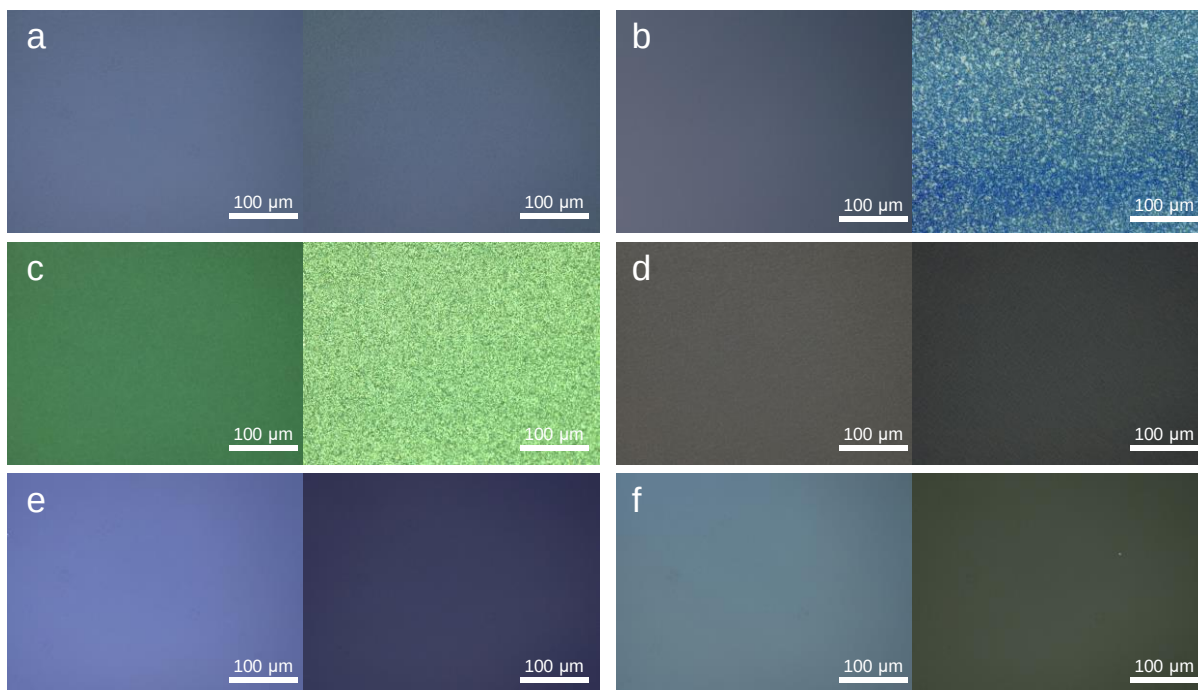
Supplementary Fig. 6 | CD and UV-vis absorption spectra of P3HT with various chiral dopants. a–e, CD spectra of (S)-BN (a), CB15 (b), R811 (c), R5011 (d), and (R, R)-TADDOL (e) doped P3HT thin films before and after thermal annealing. **f–j**, UV-vis absorption spectra of (S)-BN (f), CB15 (g), R811 (h), R5011 (i), and (R, R)-TADDOL (j) doped P3HT thin films before and after thermal annealing.



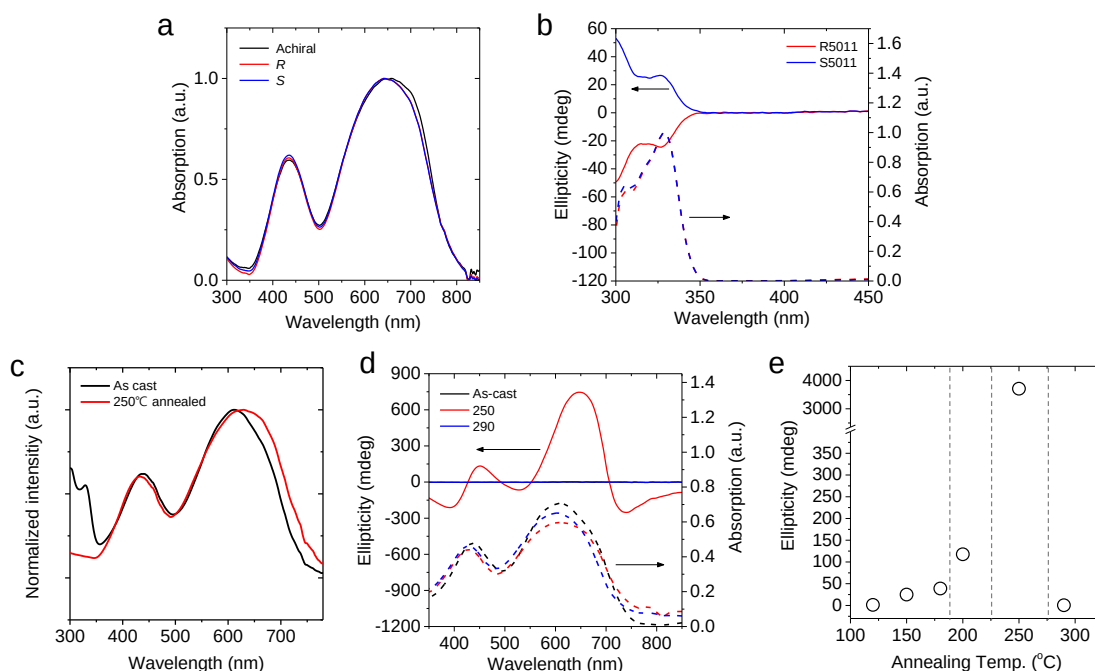
Supplementary Fig. 7 | CD and UV-vis absorption spectra of P29DPP-TTSe with various chiral dopants. a–e, CD spectra of (S)-BN (a), CB15 (b), R811 (c), S5011 (d), and (S, S)-TADDOL (e), doped PDBT-co-TT thin films before and after thermal annealing. **f–j**, UV-vis absorption spectra of (S)-BN (f), CB15 (g), R811 (h), S5011 (i), and (S, S)-TADDOL (j) doped PDBT-co-TT thin films before and after thermal annealing.



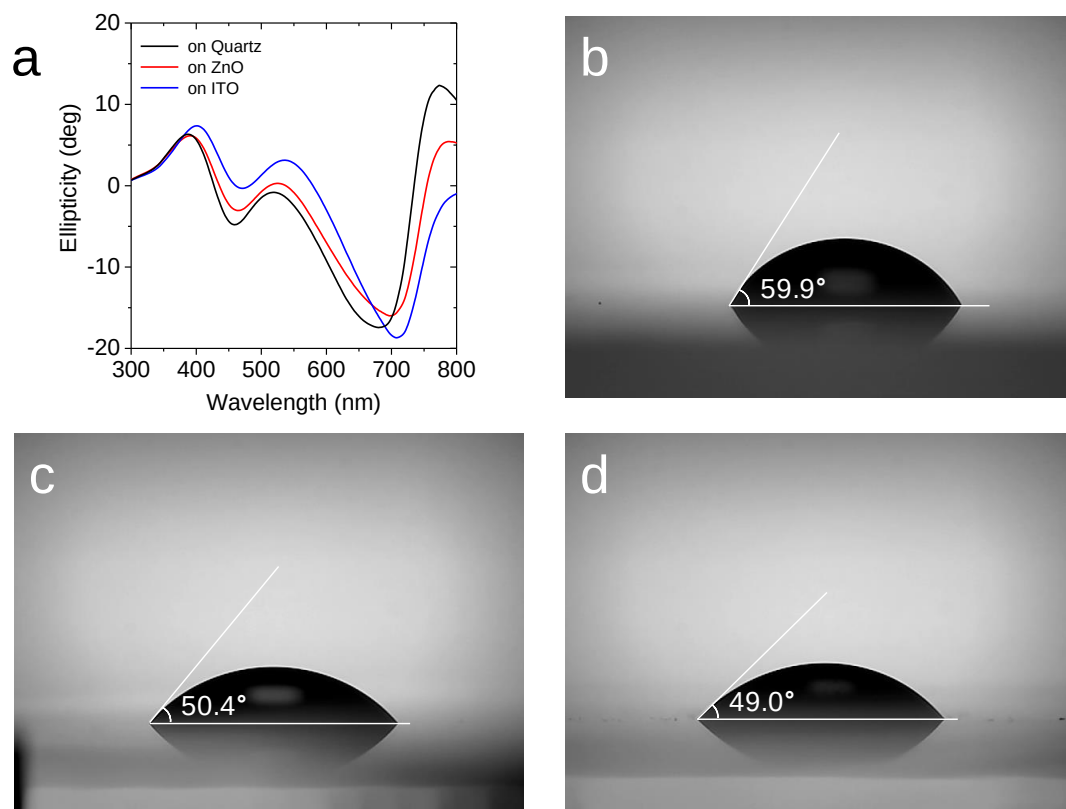
Supplementary Fig. 8 | CD and UV-vis absorption spectra of PTB7-Th with various chiral dopants. a–e, CD spectra of (S)-BN (a), CB15 (b), S811 (c), S5011 (d), and (S, S)-TADDOL (e) doped PTB7-Th thin films before and after thermal annealing. **f–j**, UV-vis absorption spectra of (S)-BN (f), CB15 (g), S811 (h), S5011 (i), and (S, S)-TADDOL (j) doped PTB7-Th thin films before and after thermal annealing.



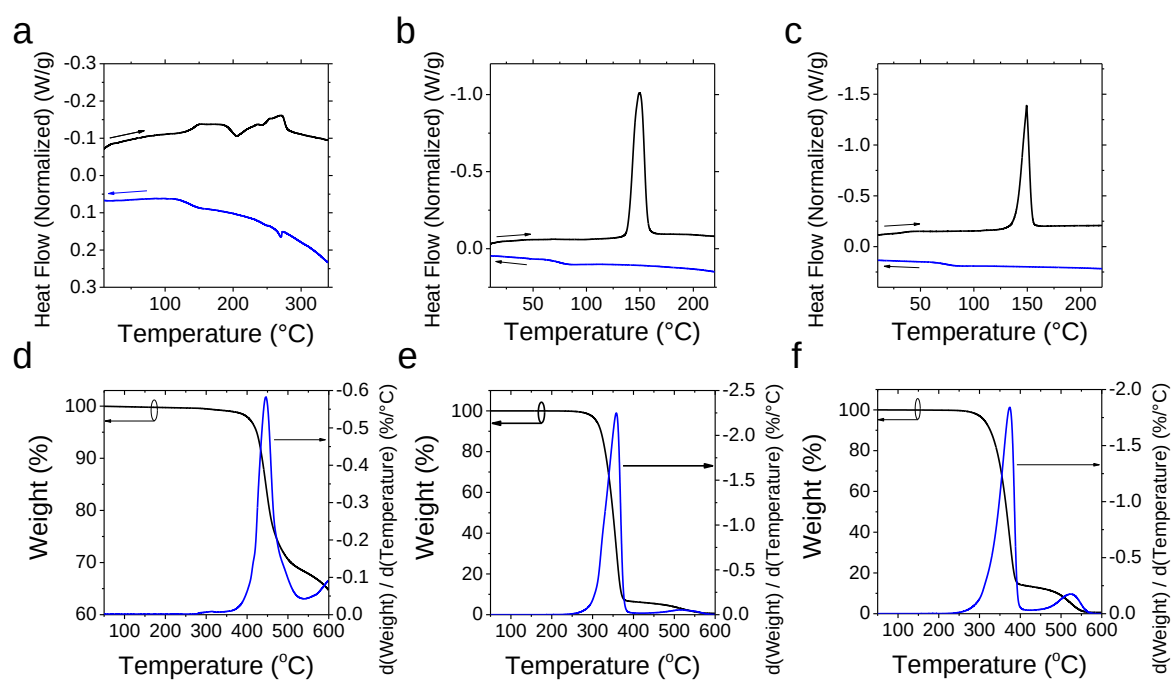
Supplementary Fig. 9 | POM images of films of the different conjugated polymers. a–f, POM images of P29DPP-TTSe (**a**), PCPDTTBTT (**b**), F8T2 (**c**), P3HT (**d**), MEH-PPV (**e**), and PTB7-Th (**f**) thin films before (left) and after (right) thermal treatment with the angle between two polarizers of 0° .



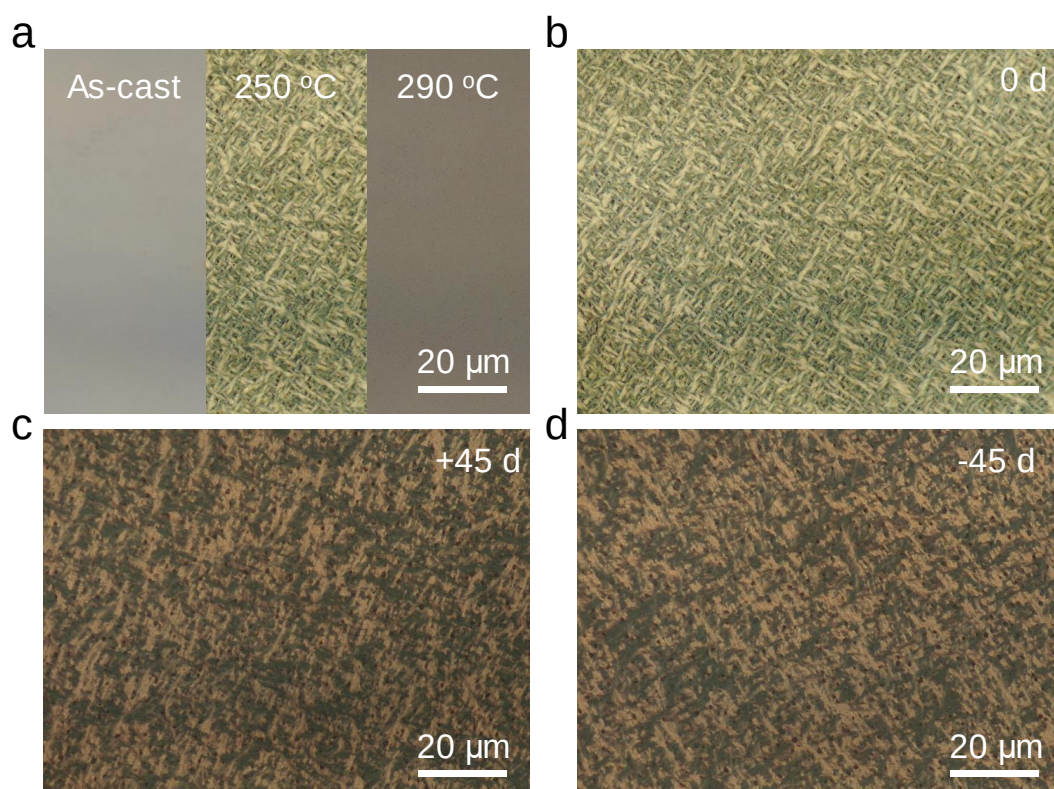
Supplementary Fig. 10 | CD and UV-vis absorption spectra of chiral active layer. **a**, UV-vis absorption spectra of CPDT-based neat and doped polymer films after thermal annealing. **b**, CD and UV-vis absorption spectra of chiral additives R5011 and S5011. **c**, UV-vis absorption spectra of CPDT-based doped polymer thin films before and after thermal annealing. **d–e**, CD and UV-vis absorption spectra (**d**) and CD intensity maxima (**e**) of CPDT-based doped thin films according to annealing temperature.



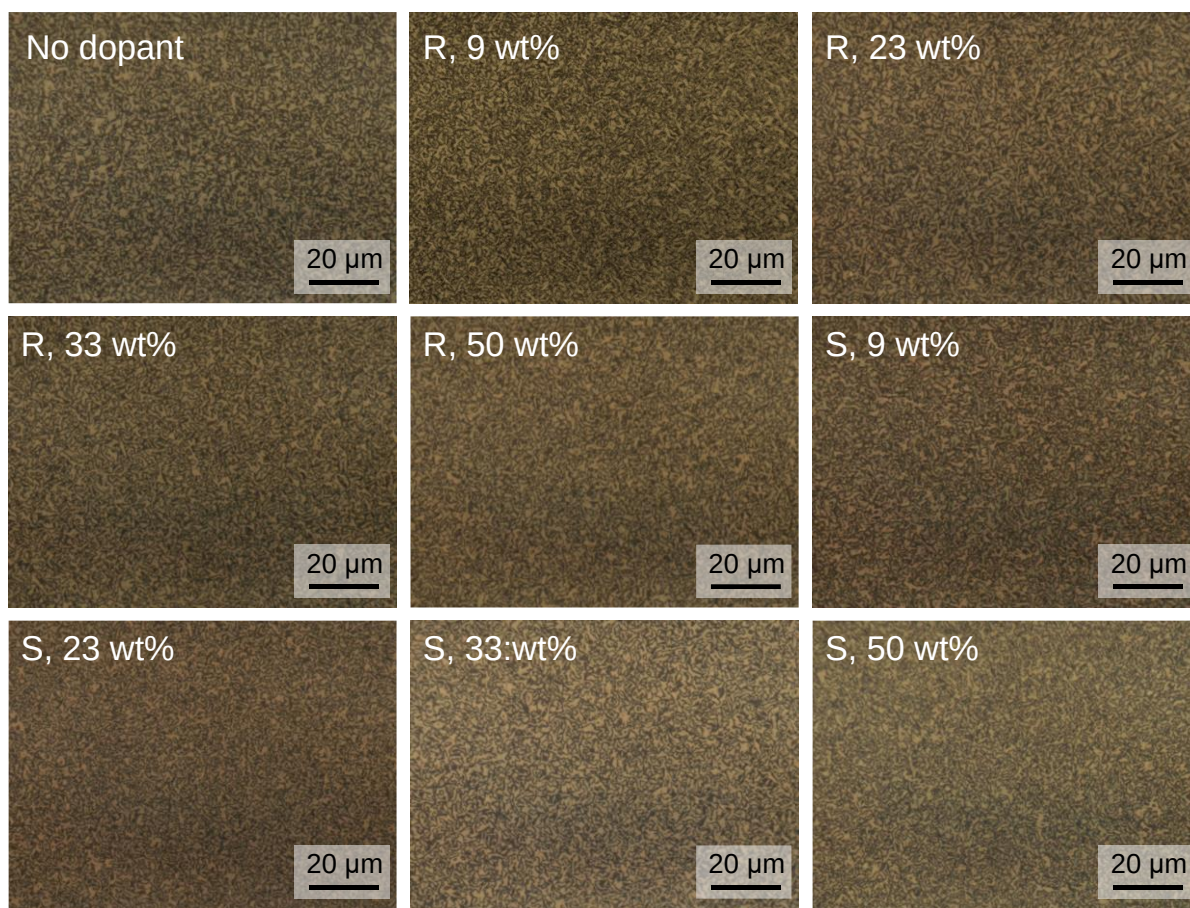
Supplementary Fig. 11 | Comparison of CD spectra for different bottom surfaces. a, CD spectra of chiral doped polymeric films spin-coated on quartz, ITO, and ZnO surfaces. **b–d**, Contact angles of water on quartz (**b**), ITO (**c**), and ZnO (**d**).



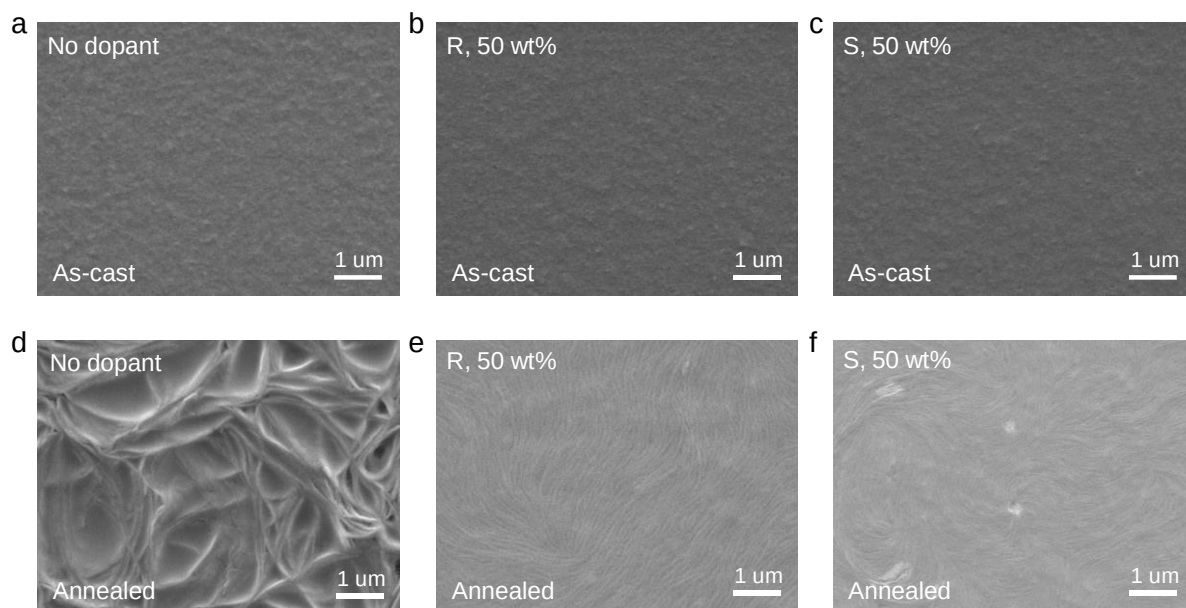
Supplementary Fig. 12 | DSC and TGA results of PCPDTTBTT and chiral dopants. a–c, DSC curves of PCPDTTBTT (a), R5011 (b), and S5011 (c) in heating (black) and cooling (blue) modes recorded at 10°C/min. **d–f,** TGA curves of PCPDTTBTT (d), R5011 (e), and S5011 (f).



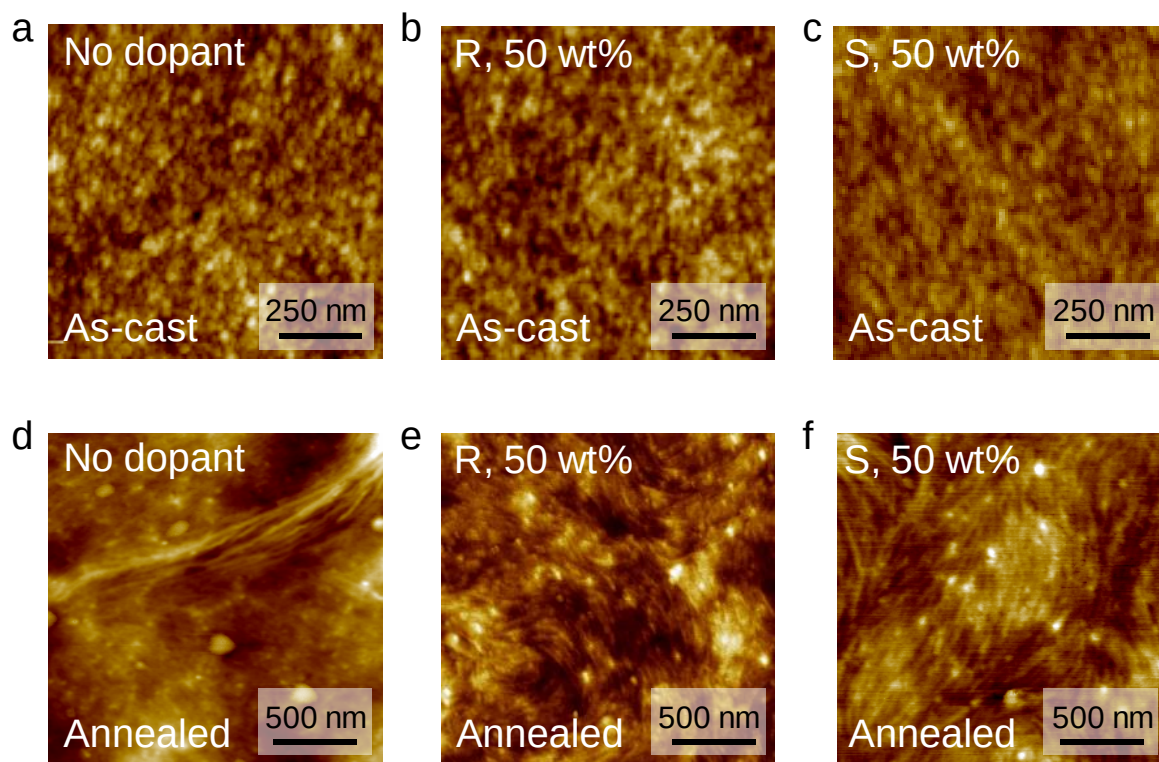
Supplementary Fig. 13 | POM images of PCPDTTBTT films. **a**, Annealing temperature-dependent POM images of PCPDTTBTT thin films. **b–d**, Images after thermal annealing with the angle between two polarizers of 0° (**b**), 45° (**c**), and –45° (**d**).



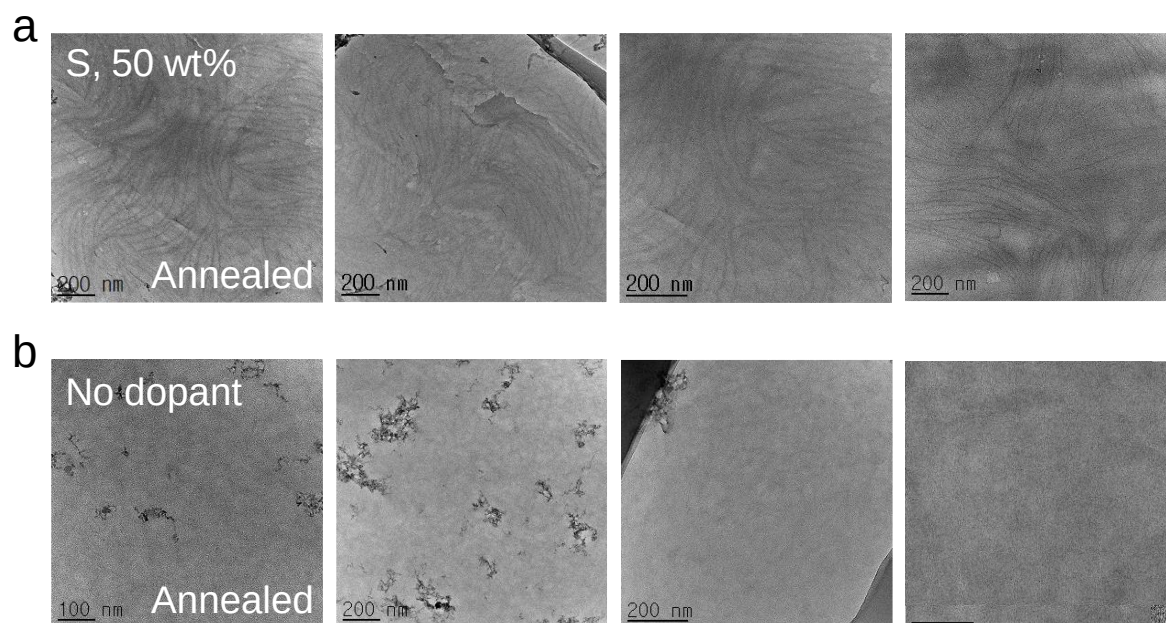
Supplementary Fig. 14 | POM images of doped polymer films after thermal treatment with various dopant concentrations.



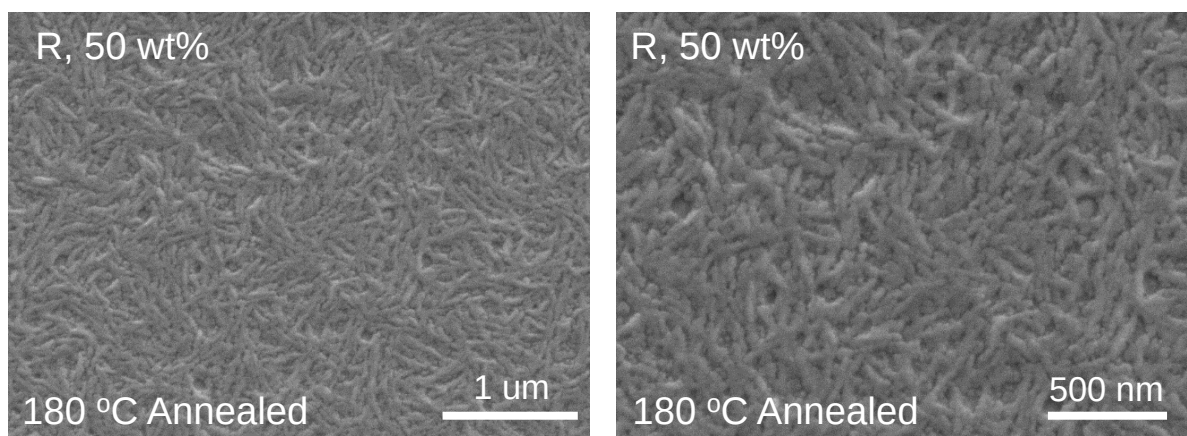
Supplementary Fig. 15 | SEM images of polymer films. **a–c**, SEM measurement results of neat PCPDTTBTT (**a**), R5011-doped (**b**), and S5011-doped (**c**) films before thermal annealing. **d–f**, SEM measurement results of neat PCPDTTBTT (**d**), R5011-doped (**e**), and S5011-doped (**f**) films after thermal annealing.



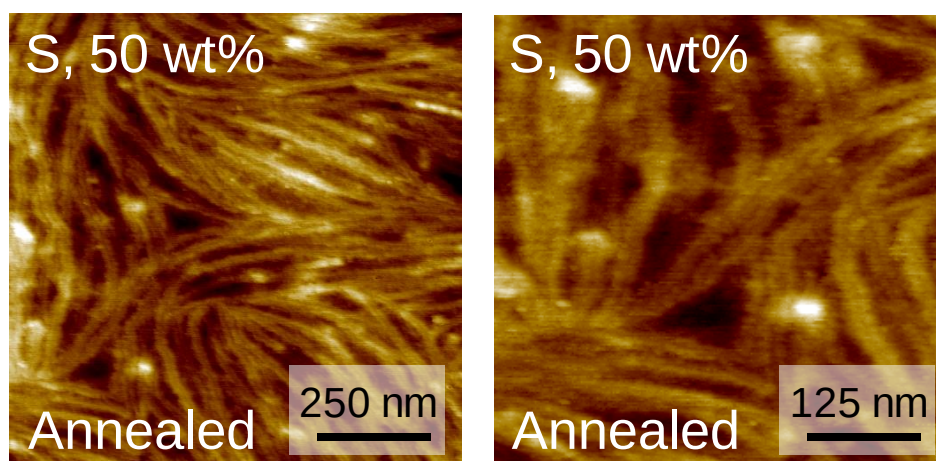
Supplementary Fig. 16 | AFM images of polymeric films. **a–c**, AFM measurement results of neat PCPDTTBTT (**a**), R5011-doped (**b**), and S5011-doped (**c**) films before thermal annealing. **d–f**, AFM measurement results of neat PCPDTTBTT (**d**), R5011-doped (**e**), and S5011-doped (**f**) films after thermal annealing.



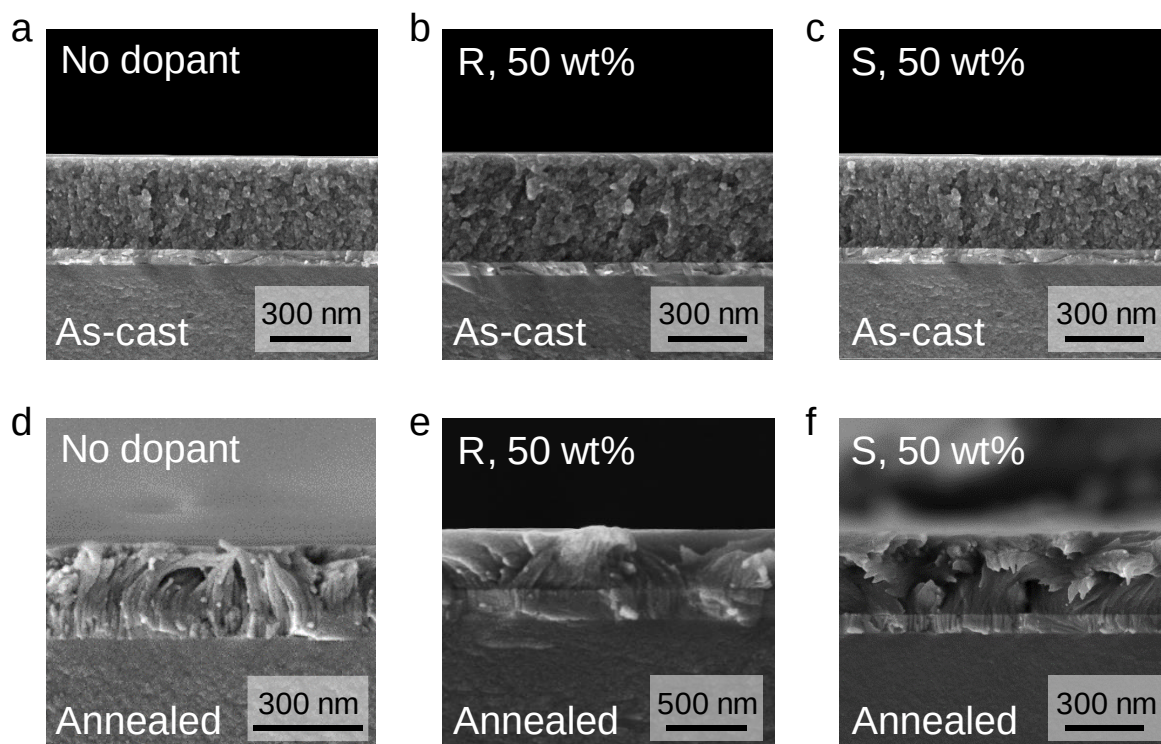
Supplementary Fig. 17 | TEM images of annealed polymeric films. a, b, TEM measurement results of S5011-doped PCPDTTBTT film (**a**) and neat PCPDTTBTT film (**b**) after thermal annealing.



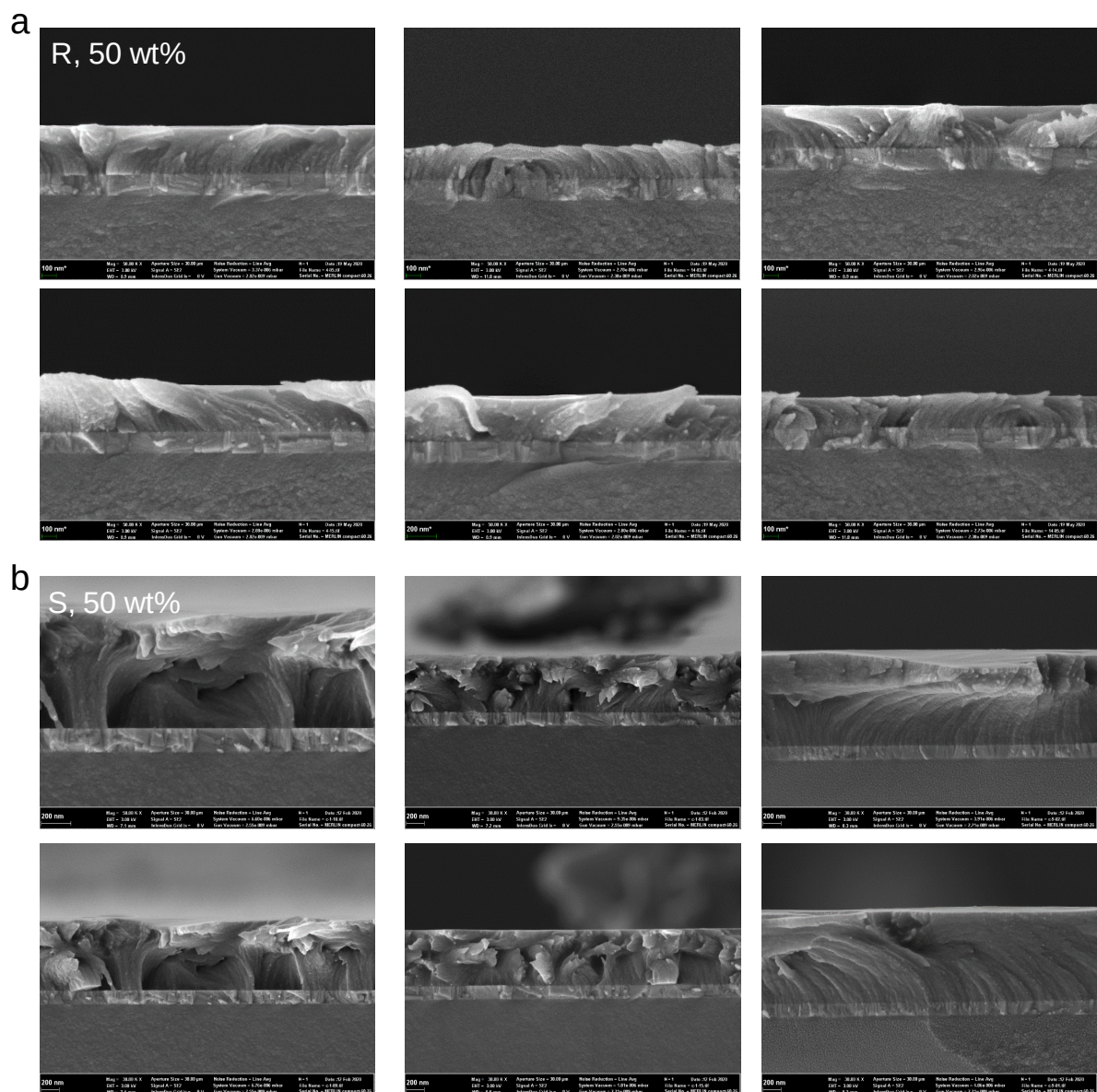
Supplementary Fig. 18 | SEM images of R5011-doped films annealed at 180°C.



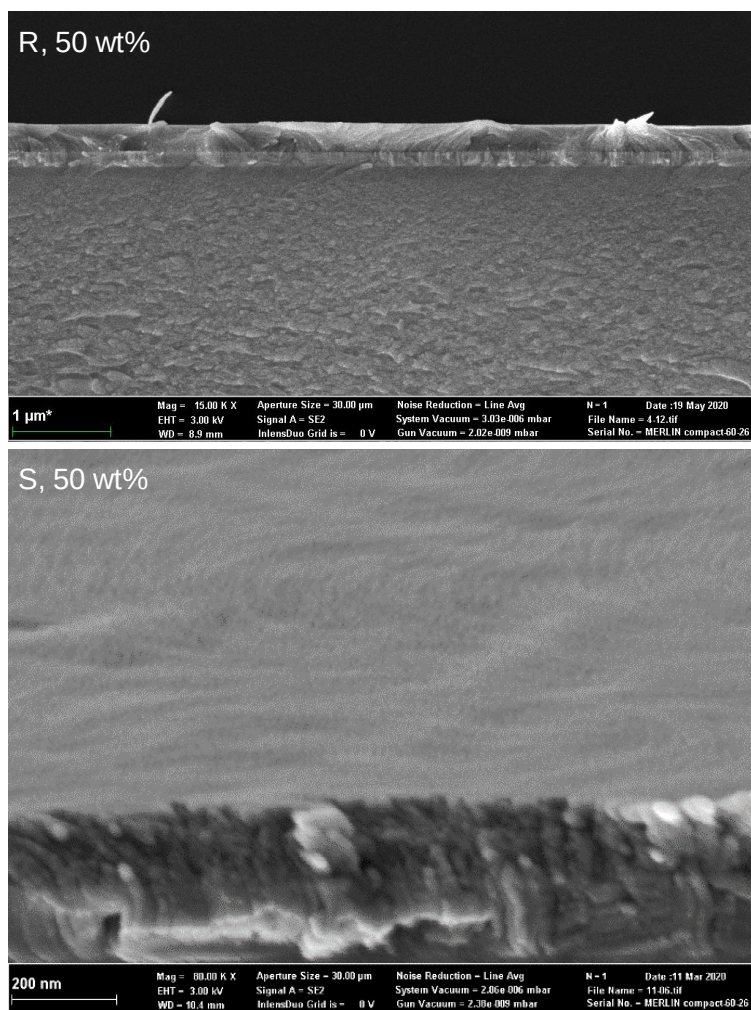
Supplementary Fig. 19 | High-resolution AFM images of S5011-doped PCPDTTBTT films after thermal annealing.



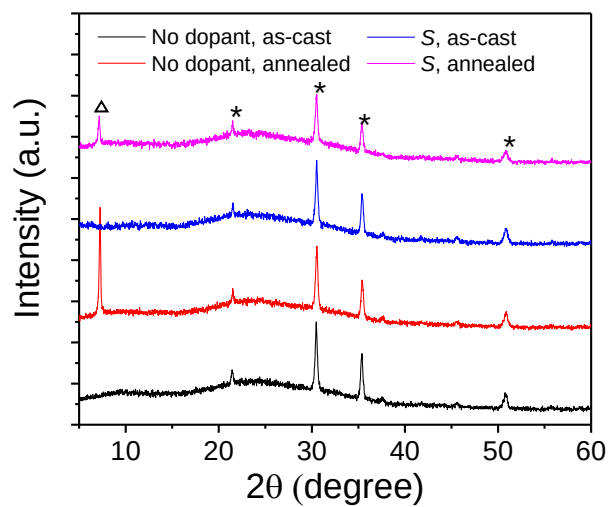
Supplementary Fig. 20 | Cross-sectional SEM images of polymeric films. **a–c**, Cross-sectional SEM measurement results of neat PCPDTTBTT (**a**), R5011-doped (**b**), and S5011-doped (**c**) films before thermal annealing. **d–f**, Cross-sectional SEM measurement results of neat PCPDTTBTT (**d**), R5011-doped (**e**), and S5011-doped (**f**) films after thermal annealing.



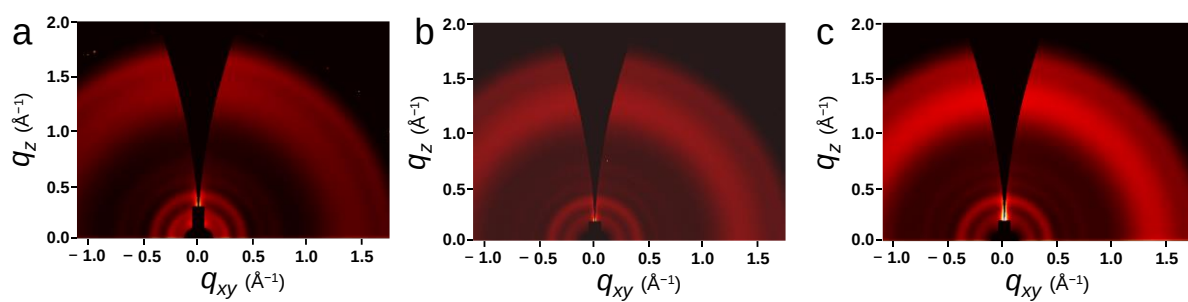
Supplementary Fig. 21 | Cross-sectional SEM images of polymeric films after thermal annealing. a, b, Cross-sectional SEM measurement results of R5011-doped PCPDTTBTT thin films (a) and S5011-doped PCPDTTBTT thin films (b) after thermal annealing.



Supplementary Fig. 22 | Large-scale and side-view cross-sectional SEM images of doped polymeric films after thermal annealing.

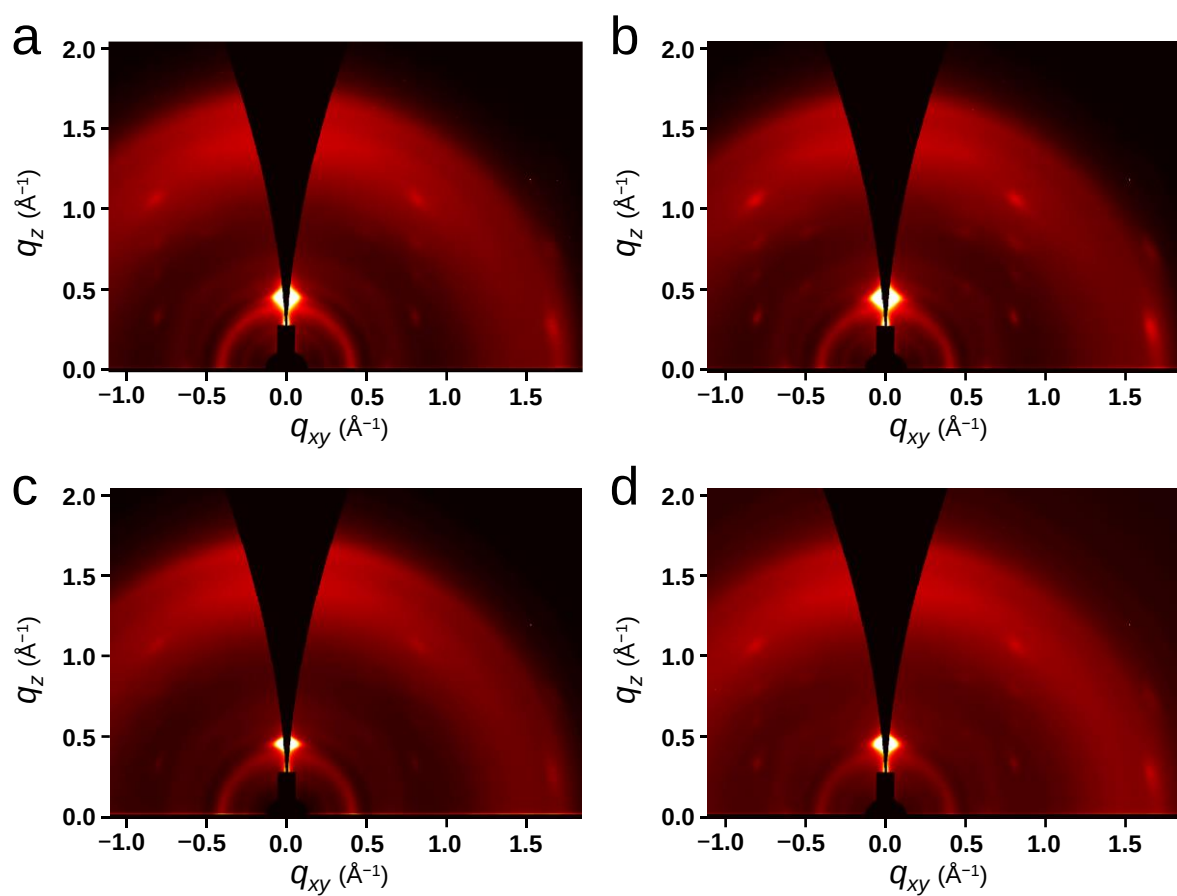


Supplementary Fig. 23 | Powder X-ray diffraction results of neat PCPDTTBTT and S5011-doped thin films before and after thermal annealing. Triangular marker represents diffraction peaks from polymers and asterisks represent diffraction peaks from zinc oxide on the substrate.



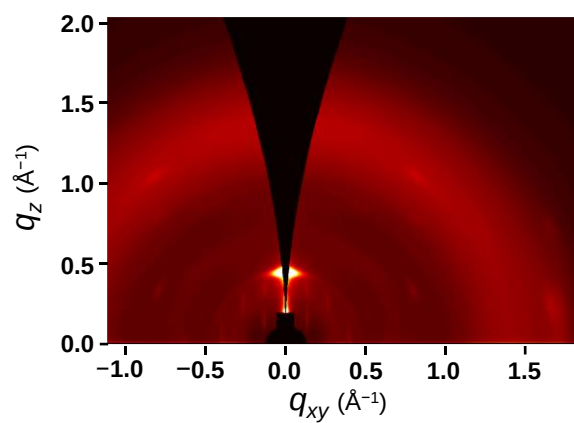
Supplementary Fig. 24 | 2D GIWAXD results of polymeric films before thermal annealing.

a–c, 2D GIWAXD measurement results of neat PCPDTTBTT (**a**), R5011-doped (**b**), and S5011-doped (**c**) films before thermal annealing with beam incident angle of 0.12° .

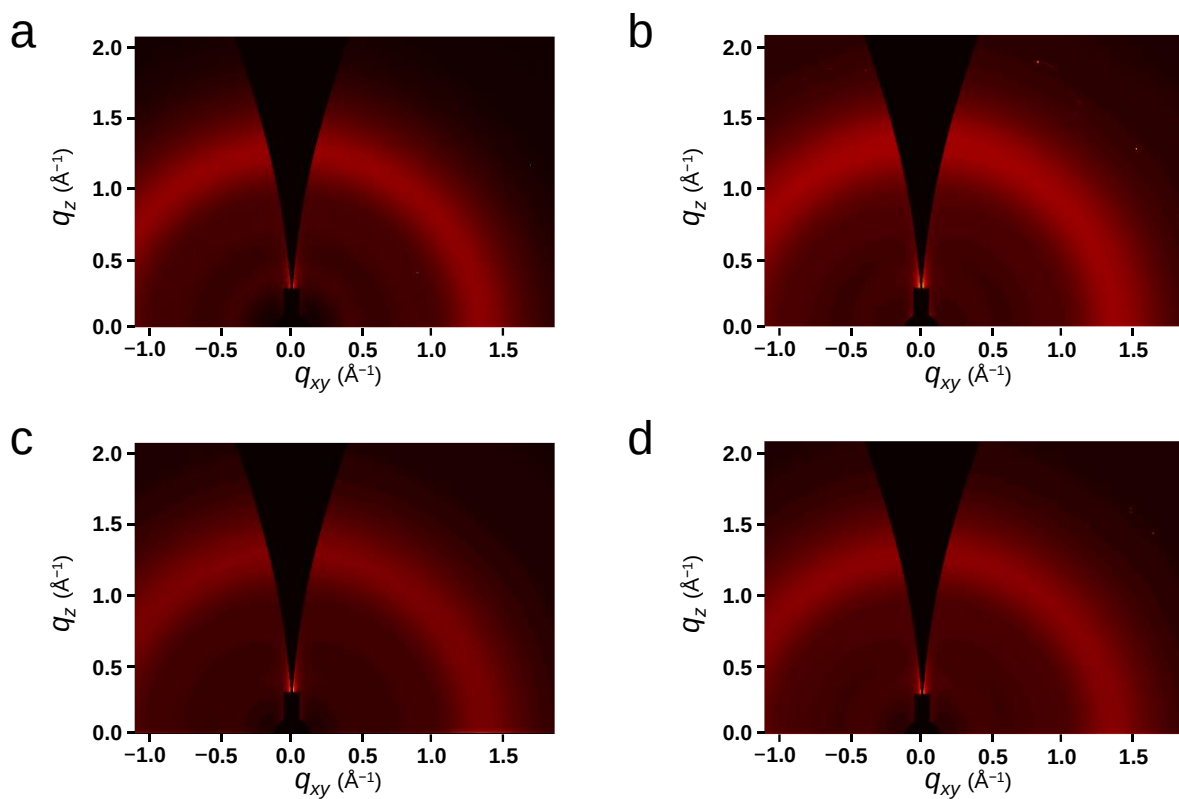


Supplementary Fig. 25 | 2D GIWAXD results of polymeric films after thermal annealing.

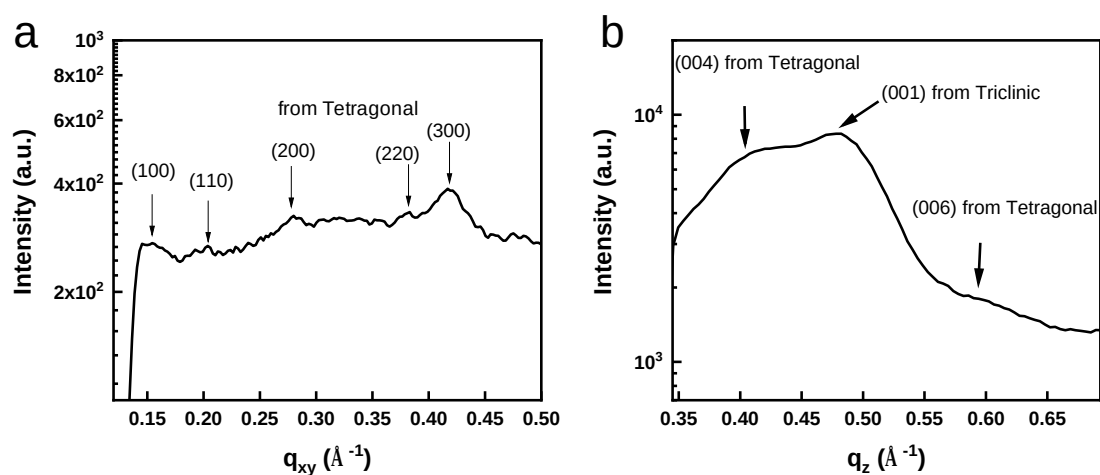
a–d, 2D GIWAXD measurement results of neat PCPDTTBTT films with beam incident angle of 0.08° (**a**), 0.1° (**b**), 0.12° (**c**), and 0.13° (**d**).



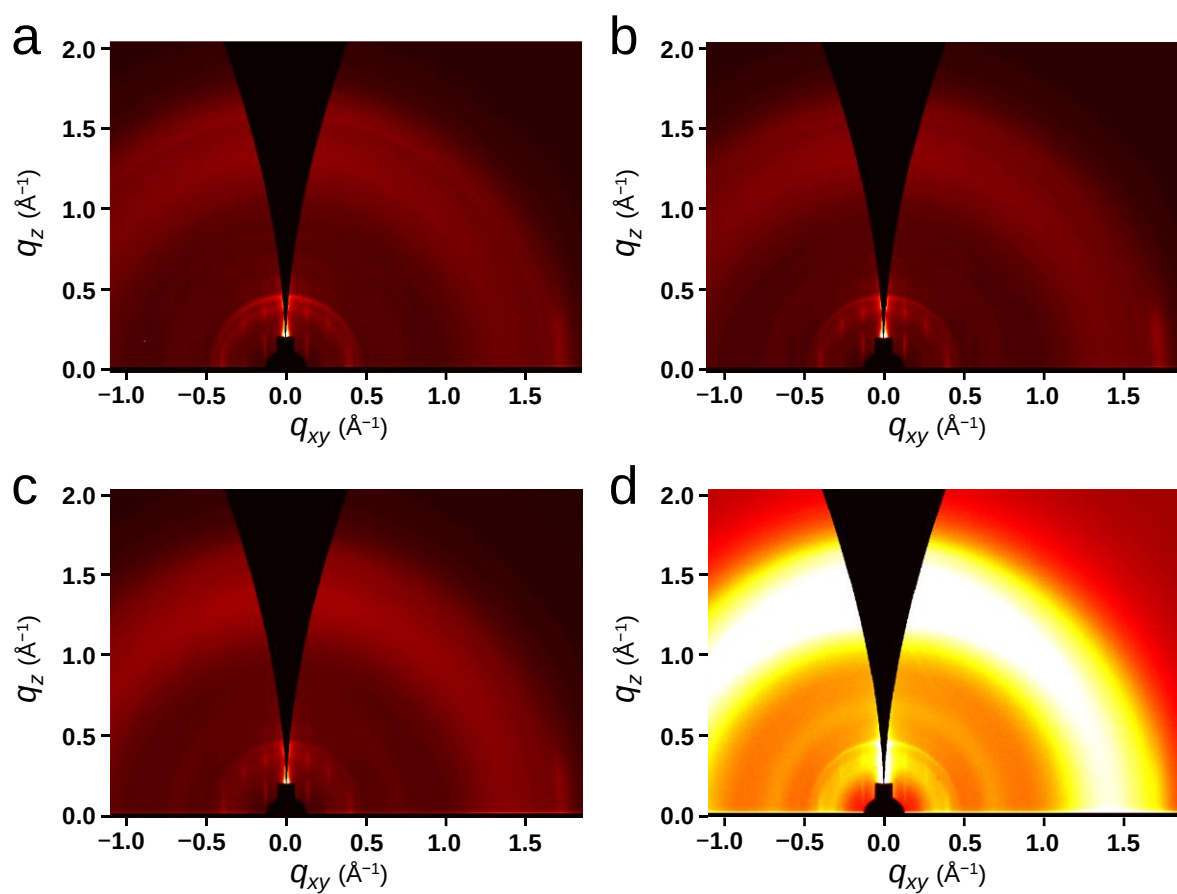
Supplementary Fig. 26 | 2D GIWAXD results of chiral doped polymeric films after thermal annealing. 2D GIWAXD measurement results of R5011-doped PCPDTTBTT films with beam incident angle of 0.12° .



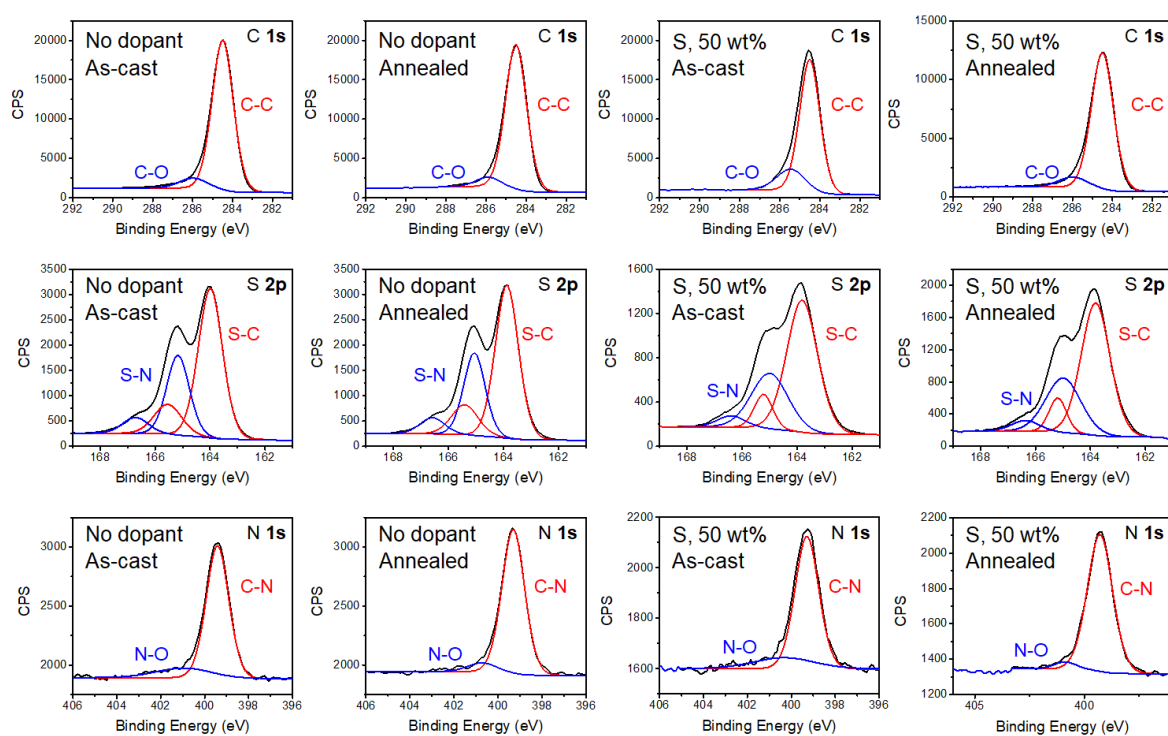
Supplementary Fig. 27 | 2D GIWAXD results of chiral dopant films. a–d, 2D GIWAXD measurement results of R5011 deposited films with beam incident angle of 0.08° (a), 0.1° (b), 0.12° (c), and 0.13° (d).



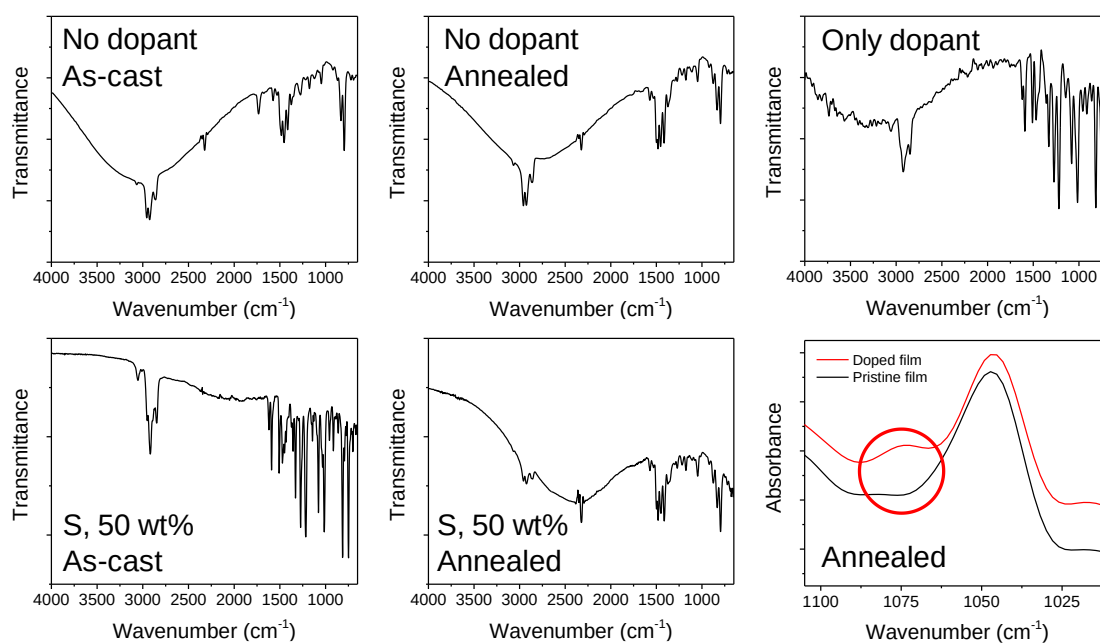
Supplementary Fig. 28 | One-dimensional GIWAXD profiles of chiral dopant films. a, b, Intensity profiles of in-plane patterns of neat polymer films (**a**) and out-of-plane patterns of R5011-doped polymer films (**b**) after chain rearrangement. Both chiral dopant films were measured with a beam incident angle of 0.12° .



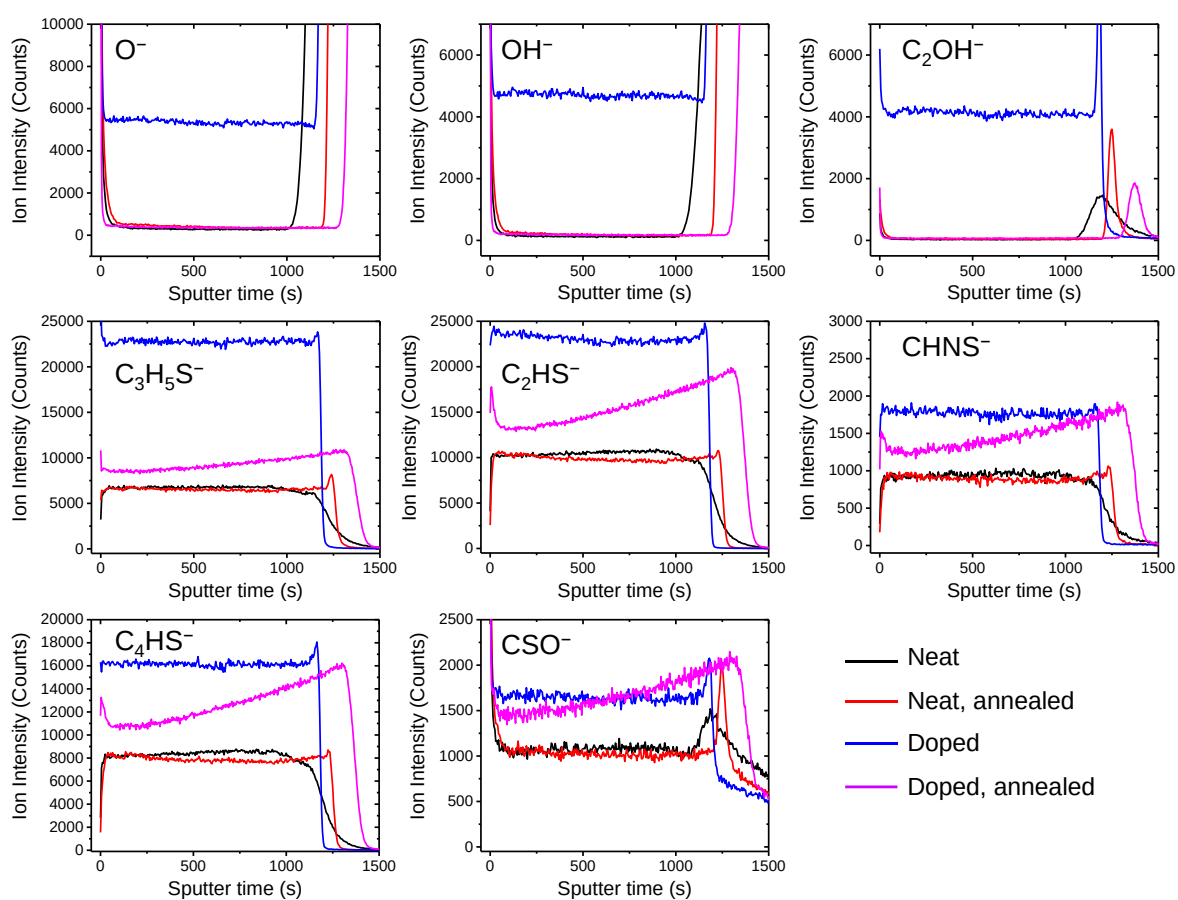
Supplementary Fig. 29 | 2D GIWAXD of chiral doped polymeric films after thermal annealing. **a–d**, 2D GIWAXD measurement results of S5011-doped PCPDTTBT films with beam incident angle of 0.08° (**a**), 0.1° (**b**), 0.12° (**c**), and 0.13° (**d**).



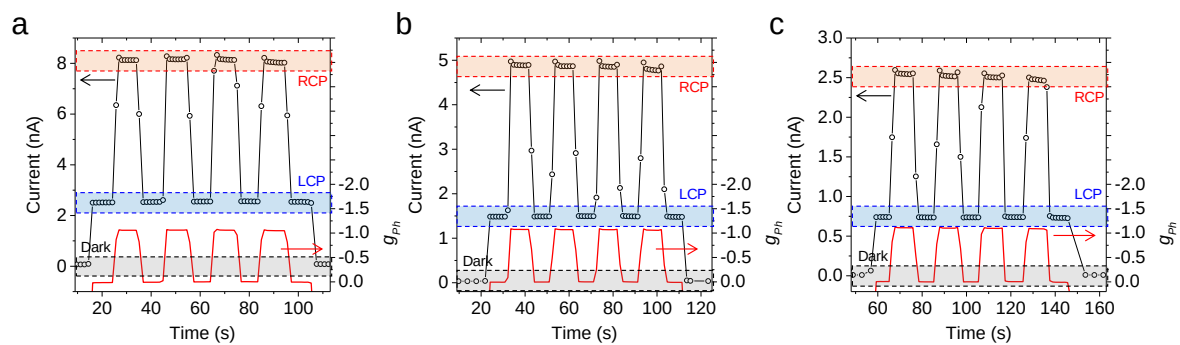
Supplementary Fig. 30 | XPS of neat PCPDTTBT and chiral doped polymeric films before and after thermal annealing.



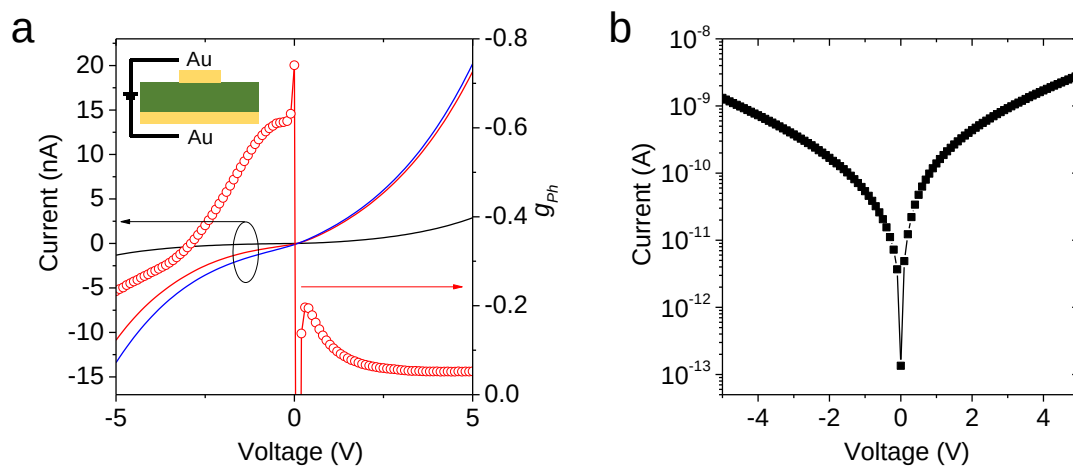
Supplementary Fig. 31 | FTIR spectra of neat PCPDTTBTT, chiral dopant, and chiral doped polymeric films according to thermal annealing.



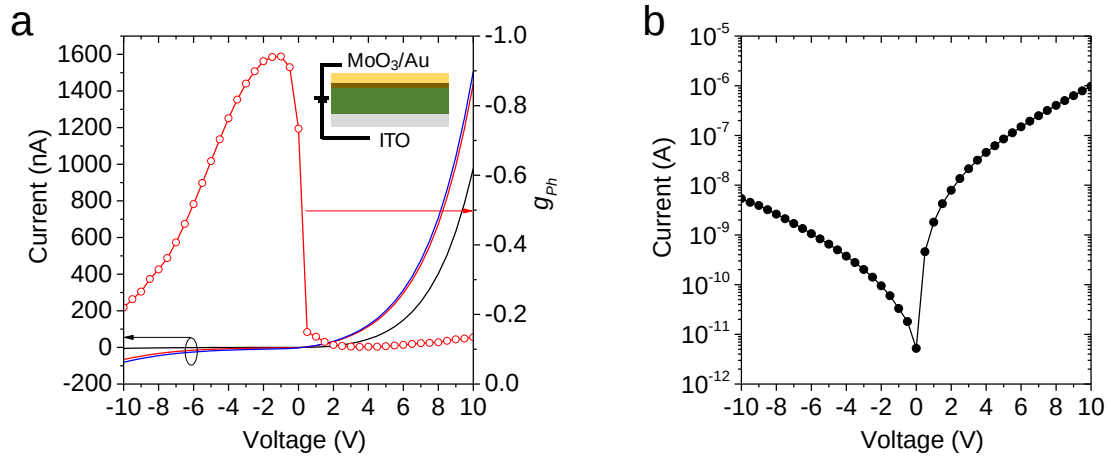
Supplementary Fig. 32 | ToF-SIMS spectra of neat PCPDTTBTT and chiral doped polymeric films before and after thermal annealing.



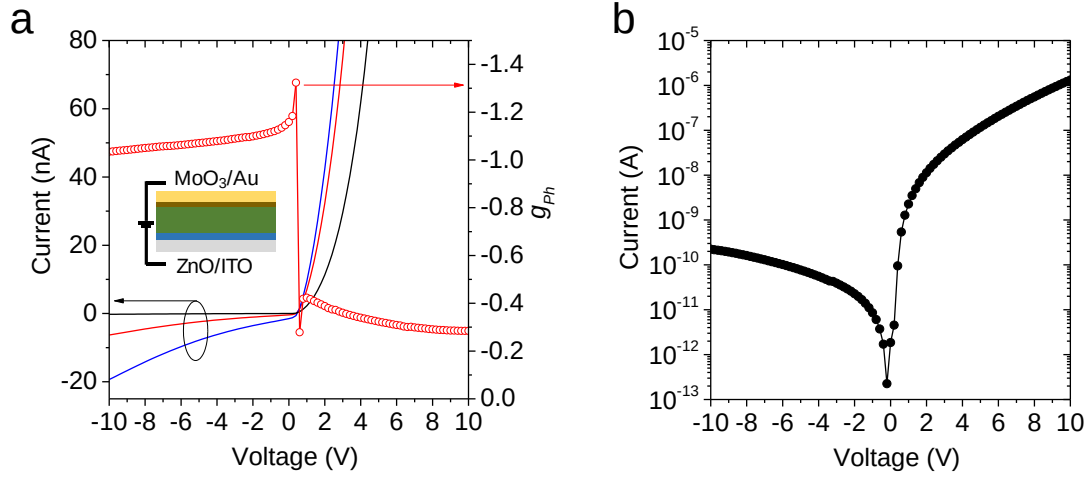
Supplementary Fig. 33 | Time-dependent SAM detection. **a–c**, Time-dependent SAM detection and corresponding photocurrent and g_{ph} values of the optimized CP photodetectors at -5 V (**a**), -3 V (**b**), and -1 V (**c**).



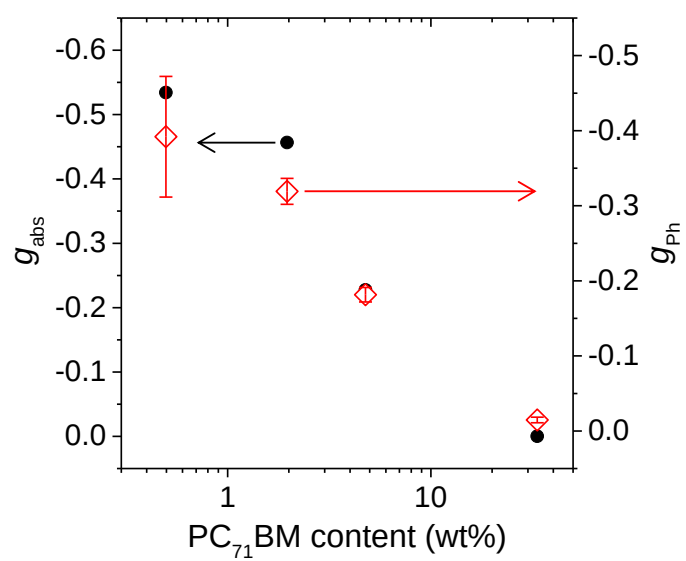
Supplementary Fig. 34 | Device characteristics of the symmetric gold/active layer/gold configuration. a, I - V characteristics and corresponding g_{Ph} values upon circular polarization detection. **b,** I - V characteristics (log scale) of photodetectors in the dark condition.



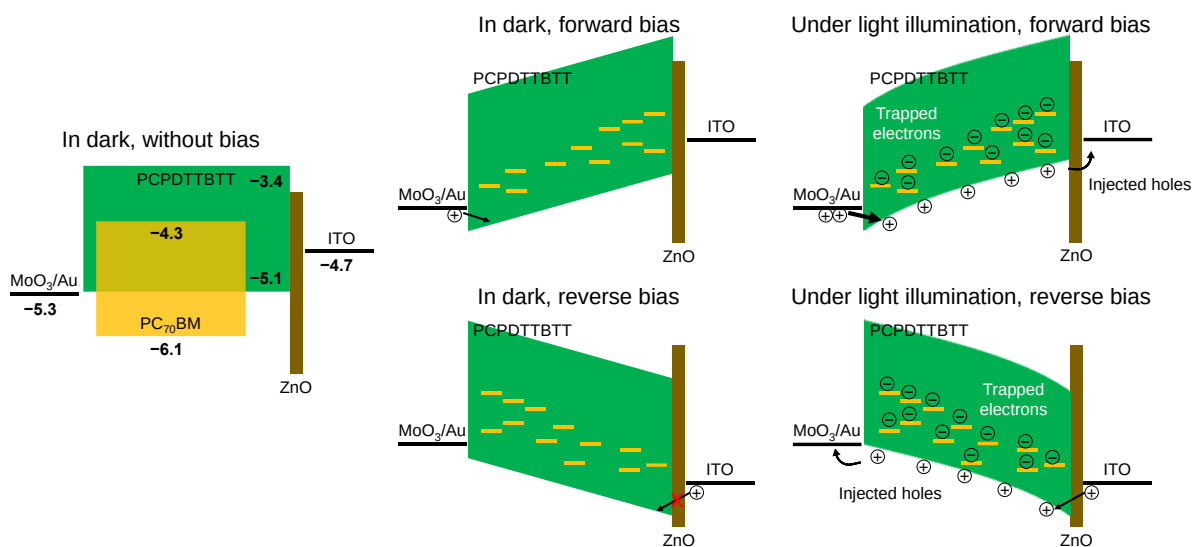
Supplementary Fig. 35 | Device characteristics of the asymmetric ITO/active layer/MoO₃-gold configuration. a, I - V characteristics and corresponding g_{ph} values upon circular polarization detection. b, I - V characteristics (log scale) of photodetectors in the dark condition.



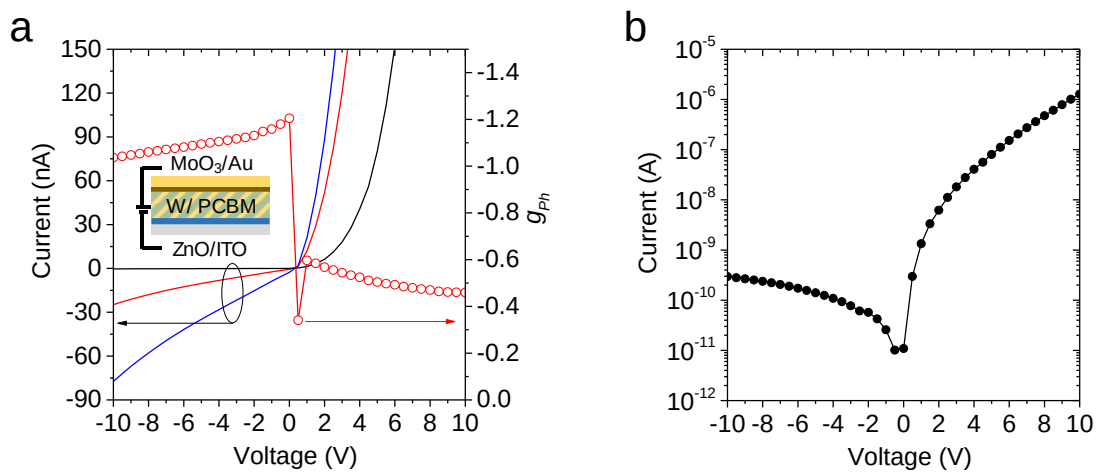
Supplementary Fig. 36 | Device characteristics of the asymmetric ITO/ZnO/active layer/MoO₃-gold configuration. a, I - V characteristics and corresponding g_{ph} values upon circular polarization detection. b, I - V characteristics (log scale) of photodetectors in the dark condition.



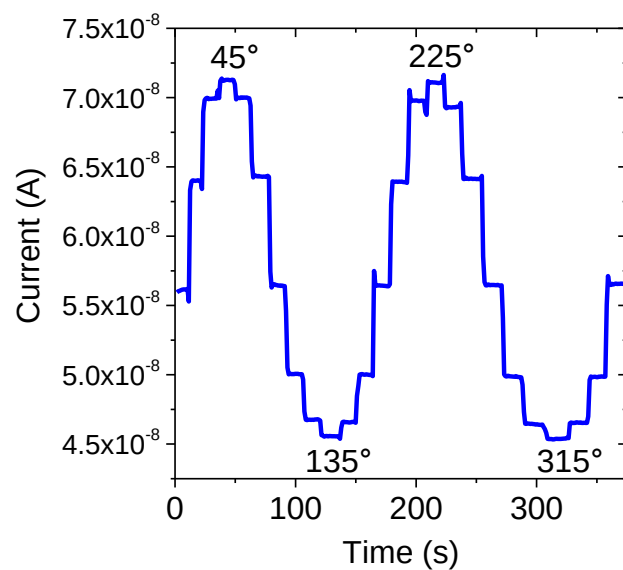
Supplementary Fig. 37 | g_{abs} and g_{ph} values of circular polarization photodetectors as a function of blended PC₇₀BM ratio.



Supplementary Fig. 38 | Schematic illustrations of the photomultiplication of charge carriers in the polymer film. Schematic illustrations of the working principles of the photomultiplication effect by interfacial trap-assisted hole tunneling injection according to the applied voltage and light illumination.

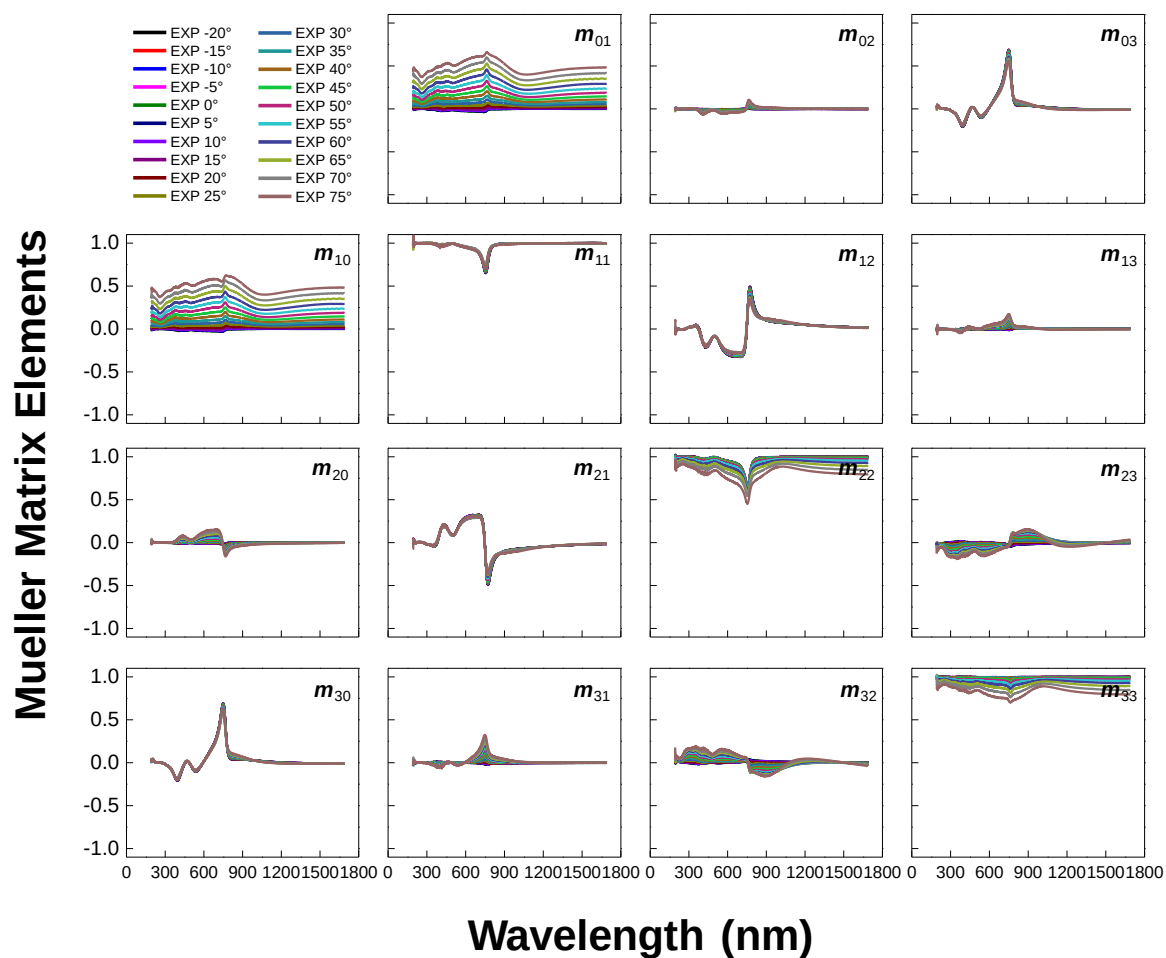


Supplementary Fig. 39 | Device characteristics of the asymmetric ITO/ZnO/active layer (with 1 wt% PC₇₀BM)/gold configuration. a, I - V characteristics and corresponding g_{ph} values upon circularly polarized light detection. b, I - V characteristics (log scale) of photodetectors in the dark condition.

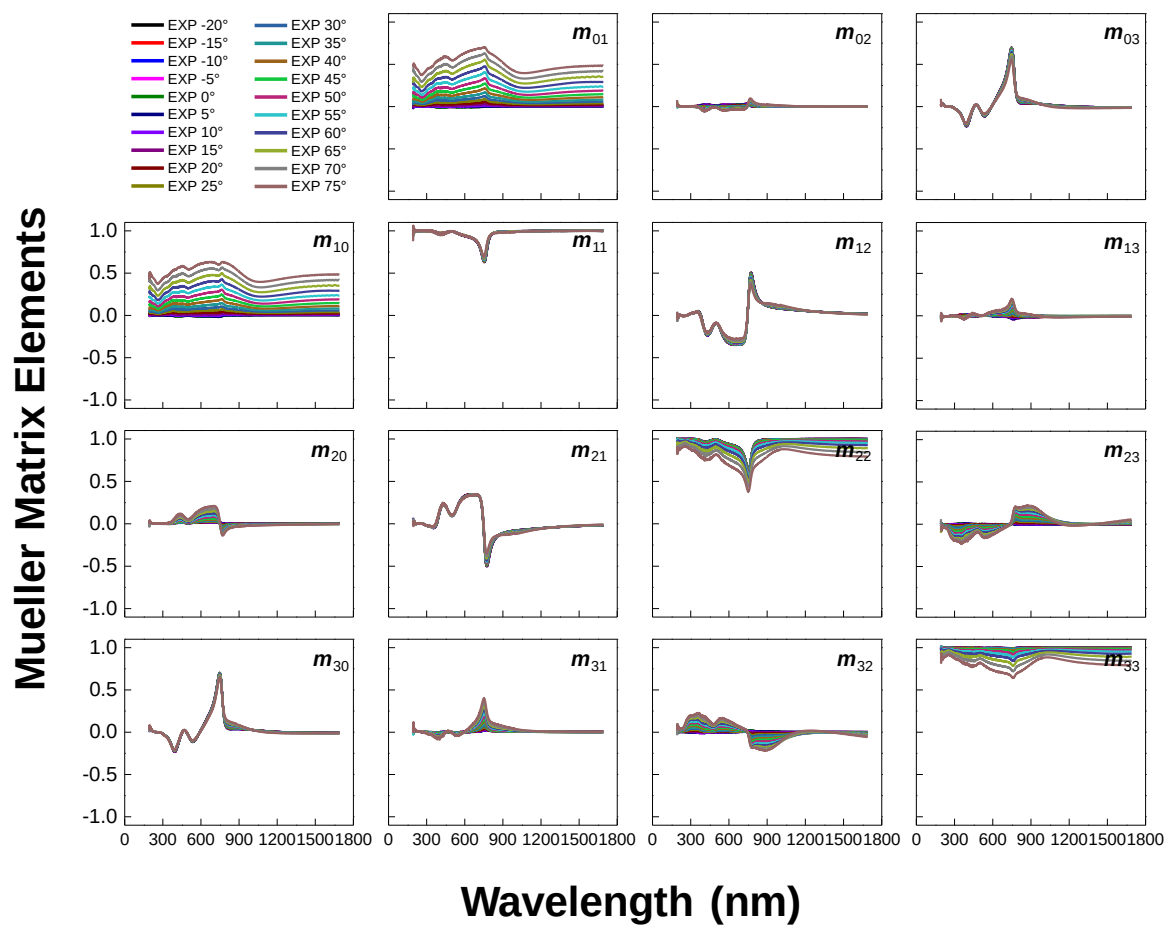


Supplementary Fig. 40 | Dynamic degree of circular polarization measurement results.

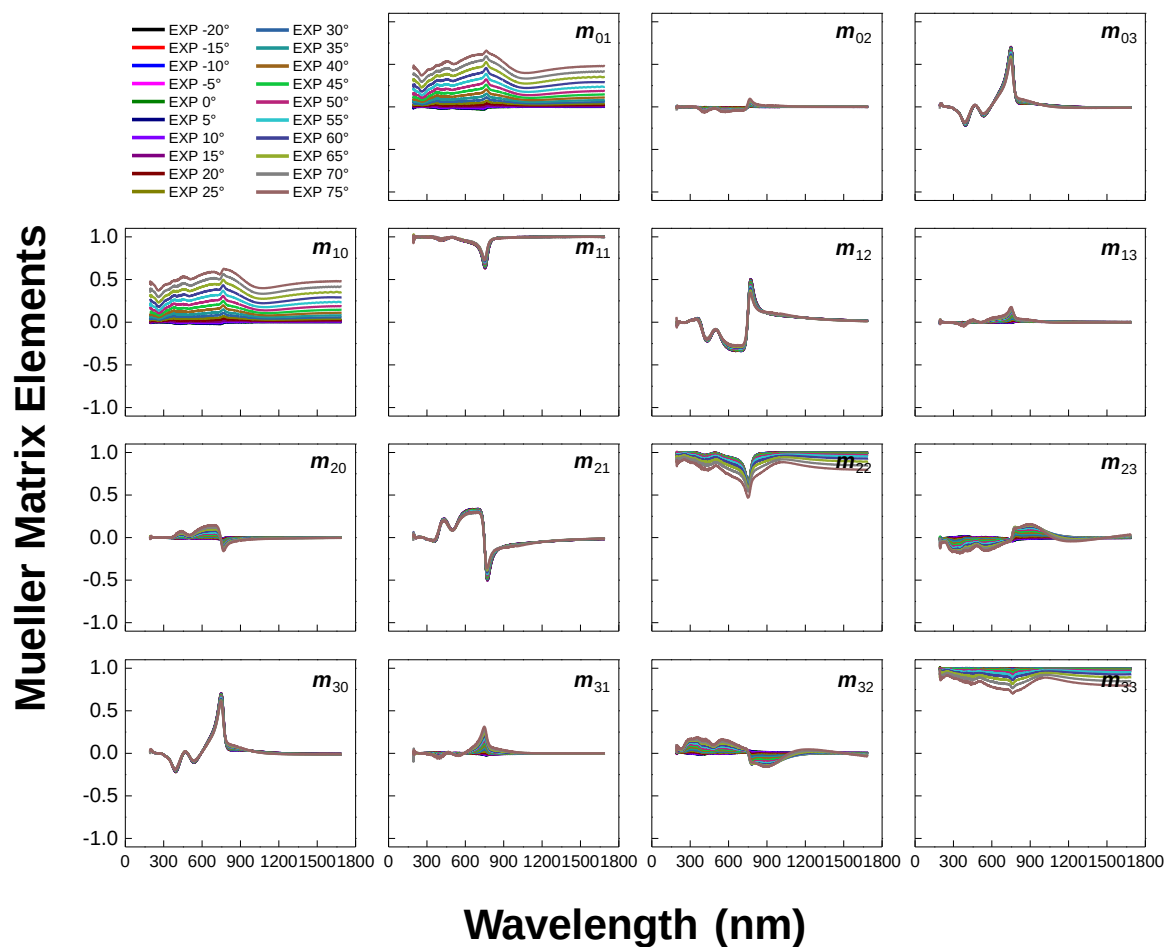
Time-dependent photocurrents of circular polarization photodetectors were measured by rotating the quarter-wave plate in 15° increments for polarized beam generation. The angle between the linear polarizer and quarter-wave plate in the first region was 0°.



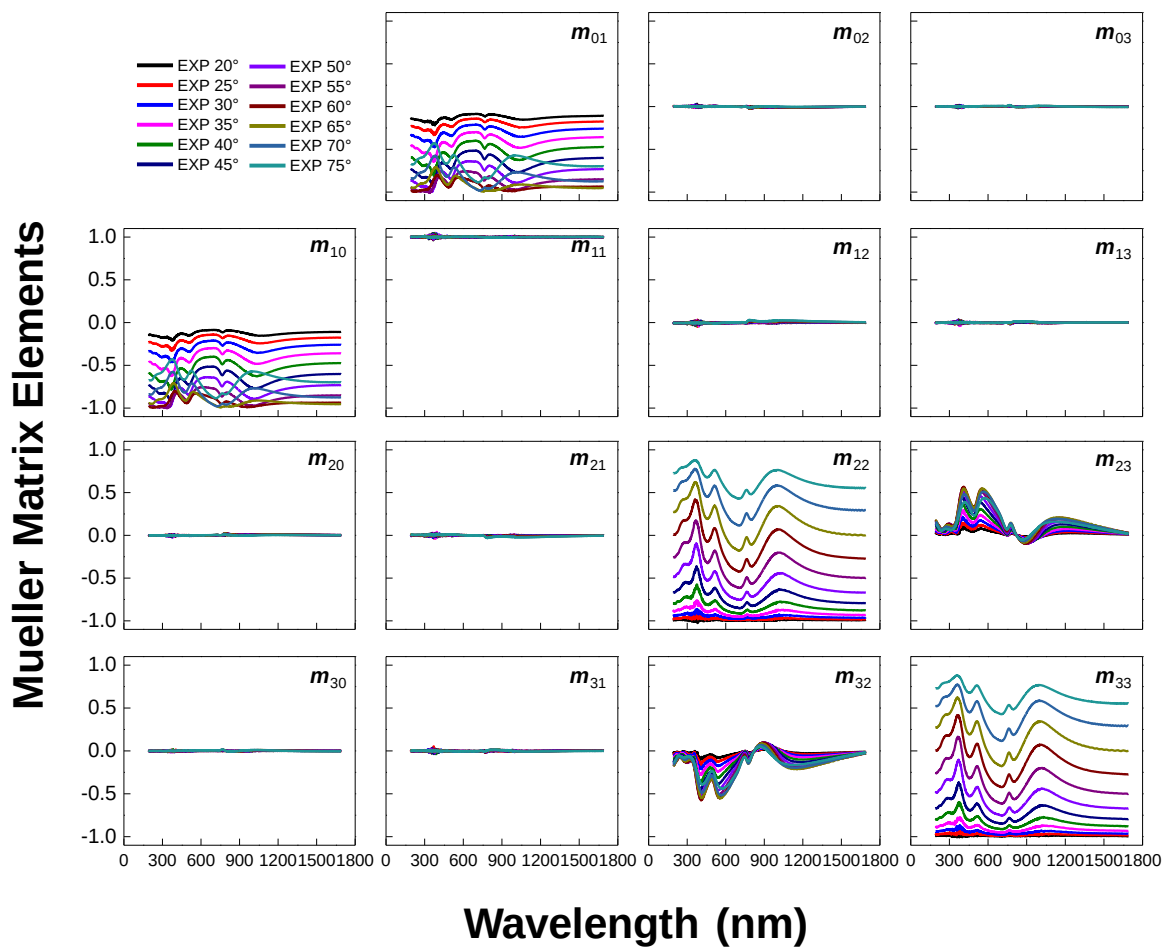
Supplementary Fig. 41 | MMSE spectra for various contributions to optical activity for the R5011-doped PCPDTTBT film. MMSE data were recorded in transmission mode with 0° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



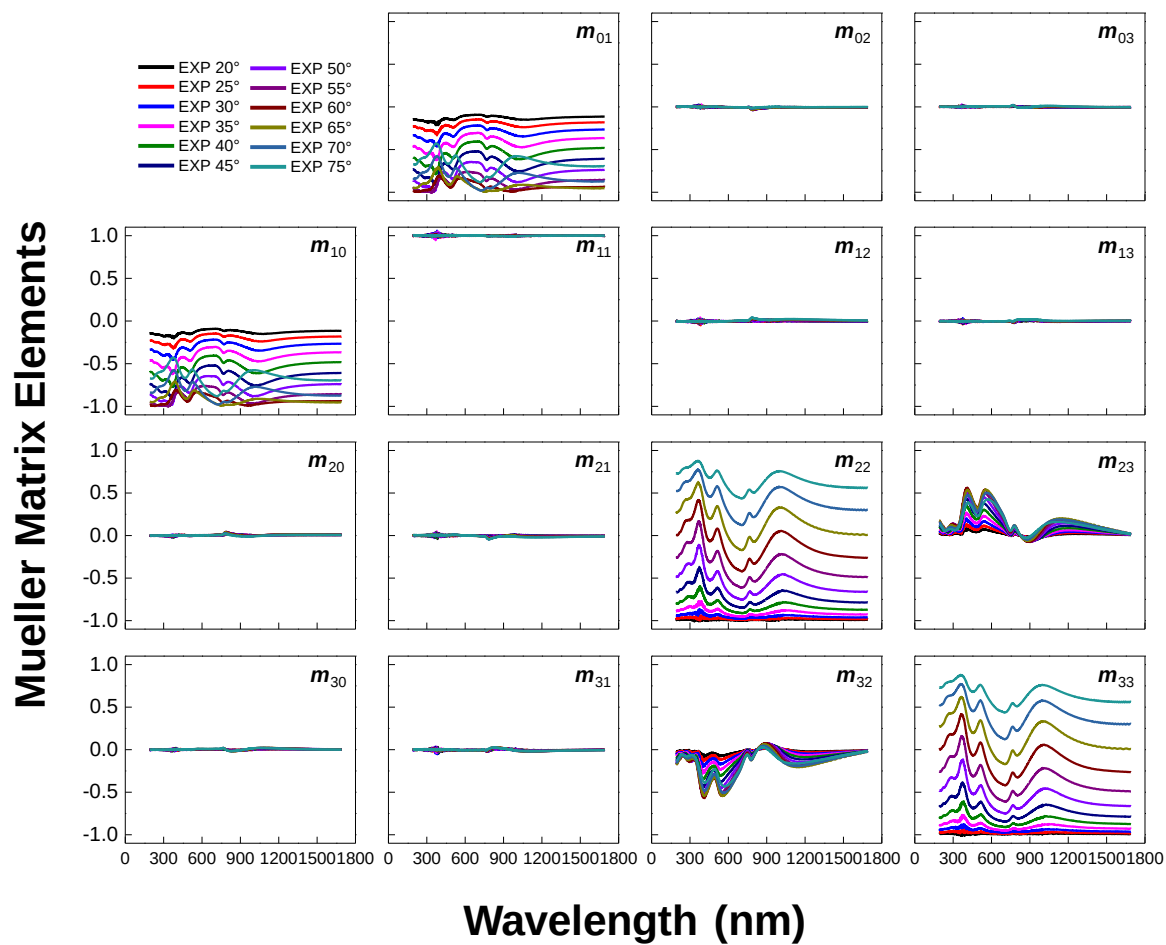
Supplementary Fig. 42 | MMSE for various contributions to optical activity for the R5011-doped PCPDTTBT film. MMSE data were recorded in transmission mode with 45° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



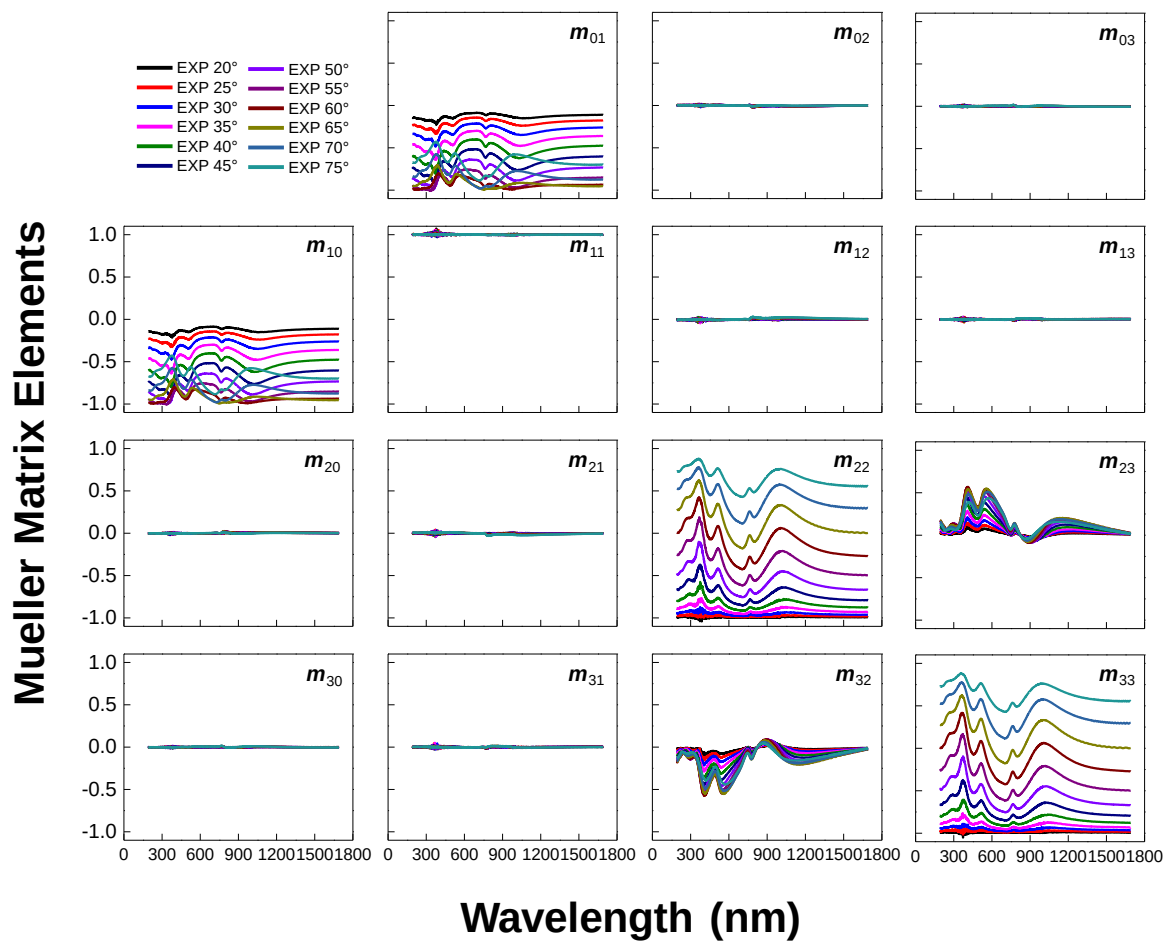
Supplementary Fig. 43 | MMSE for various contributions to optical activity for the R5011-doped PCPDTTBTT film. MMSE data were recorded in transmission mode with 90° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



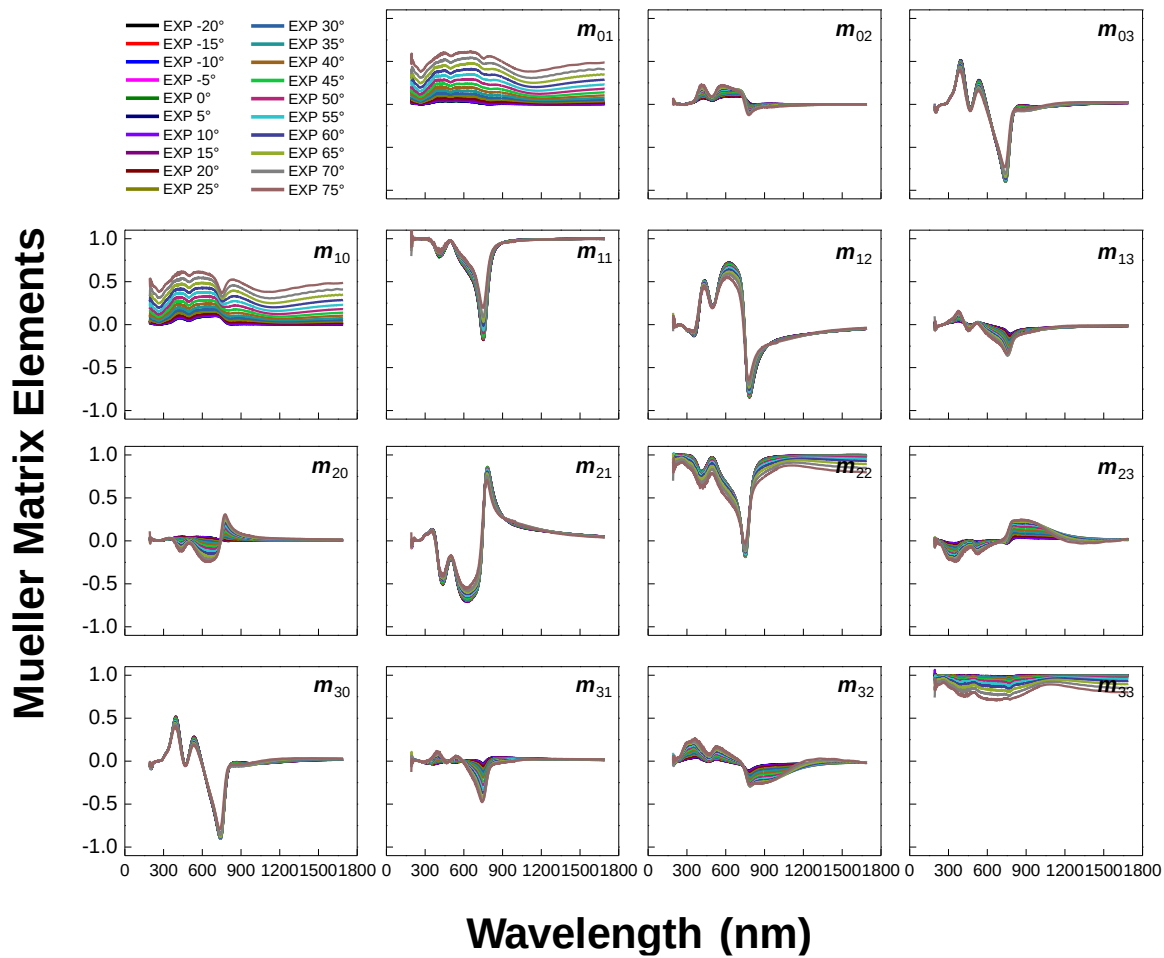
Supplementary Fig. 44 | MMSE for various contributions to optical activity for the R5011-doped PCPDTTBTT film. MMSE data were recorded in reflection mode with 0° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



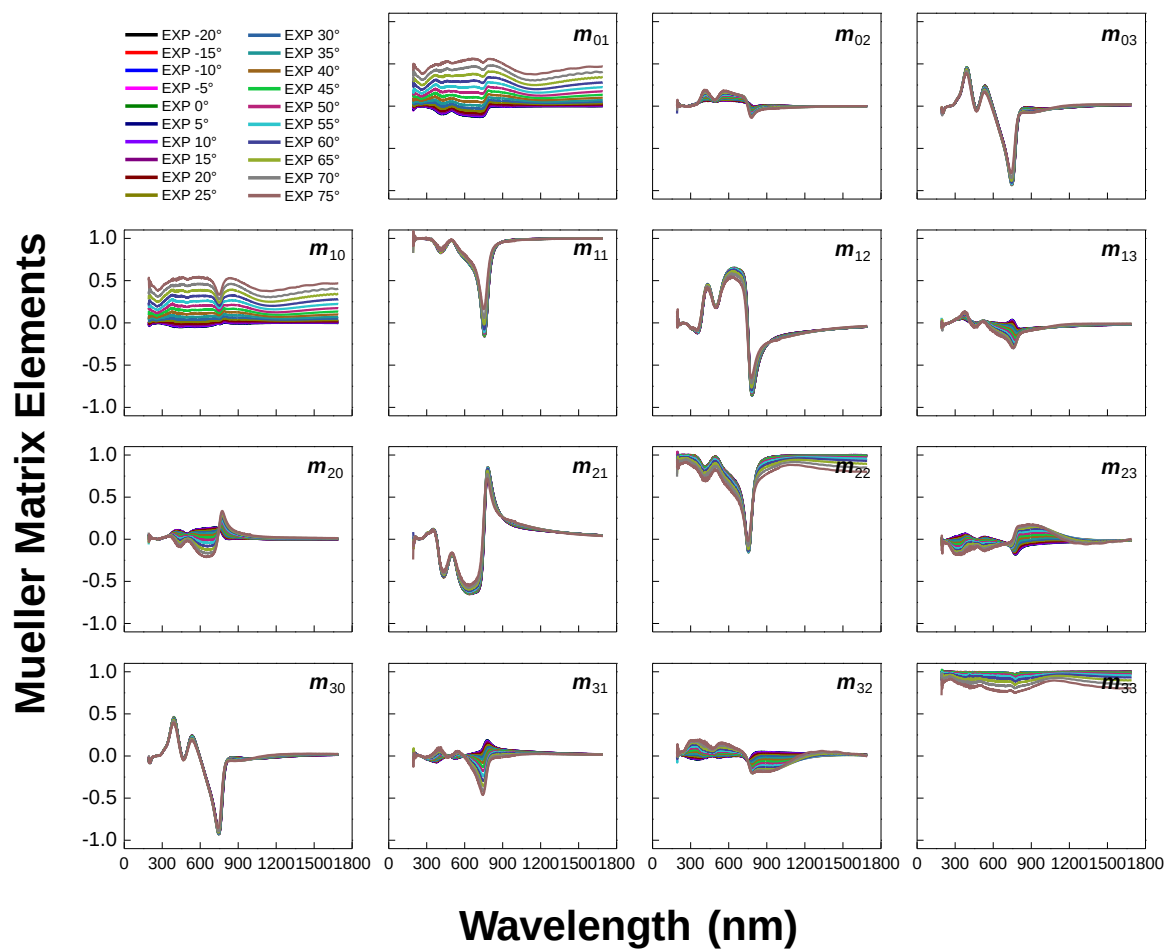
Supplementary Fig. 45 | MMSE for various contributions to optical activity for the R5011-doped PCPDTTBTT film. MMSE data were recorded in reflection mode with 45° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



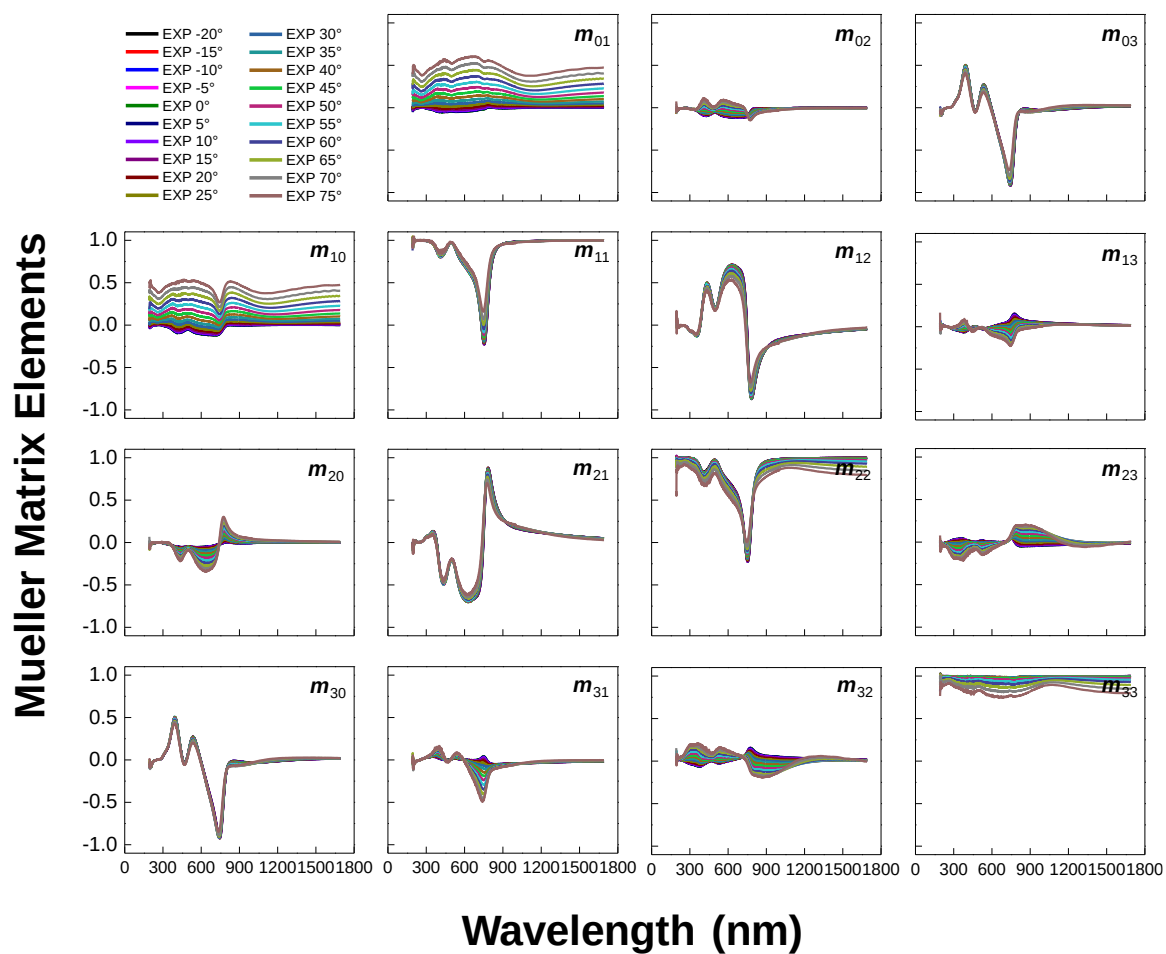
Supplementary Fig. 46 | MMSE for various contributions to optical activity for the R5011-doped PCPDTTBT film. MMSE data were recorded in reflection mode with 90° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



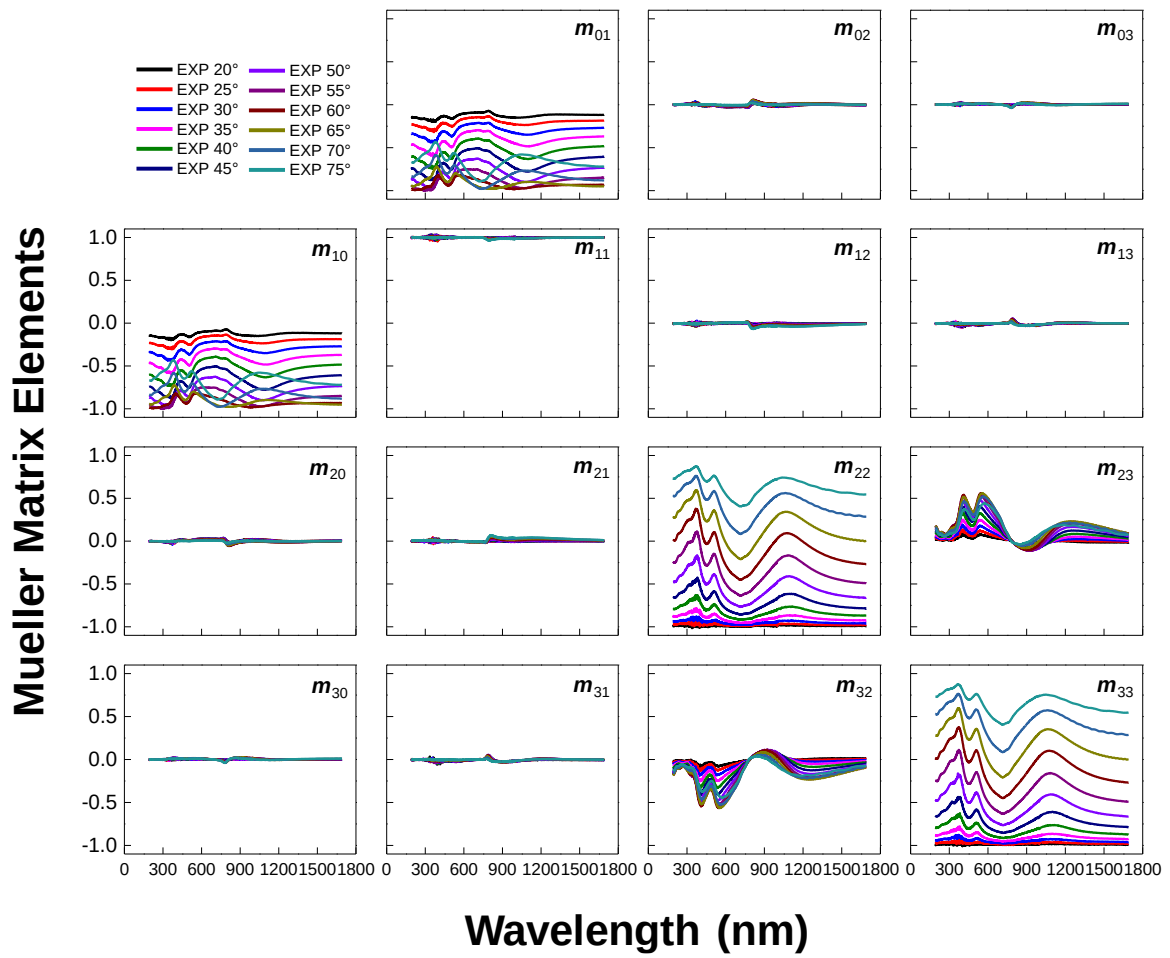
Supplementary Fig. 47 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in transmission mode with 0° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



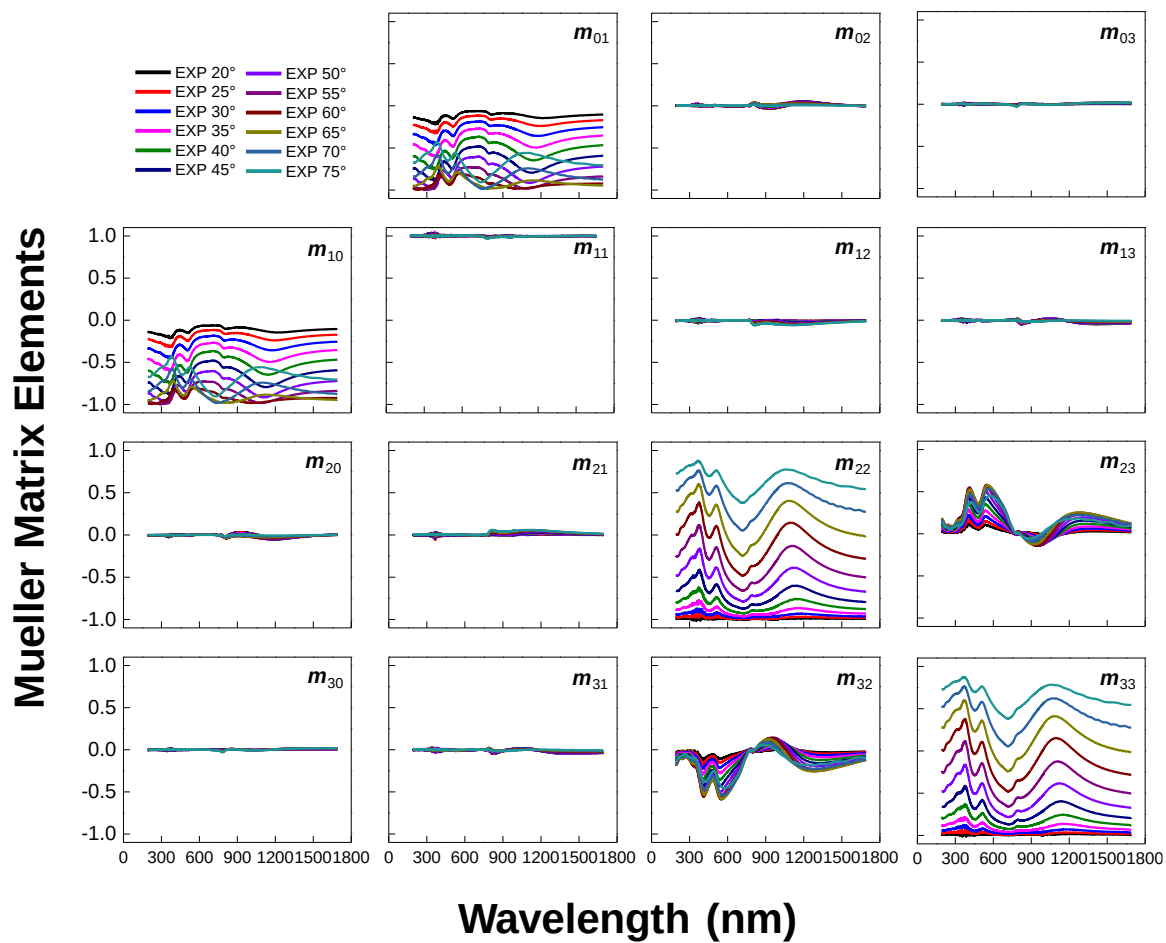
Supplementary Fig. 48 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in transmission mode with 45° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



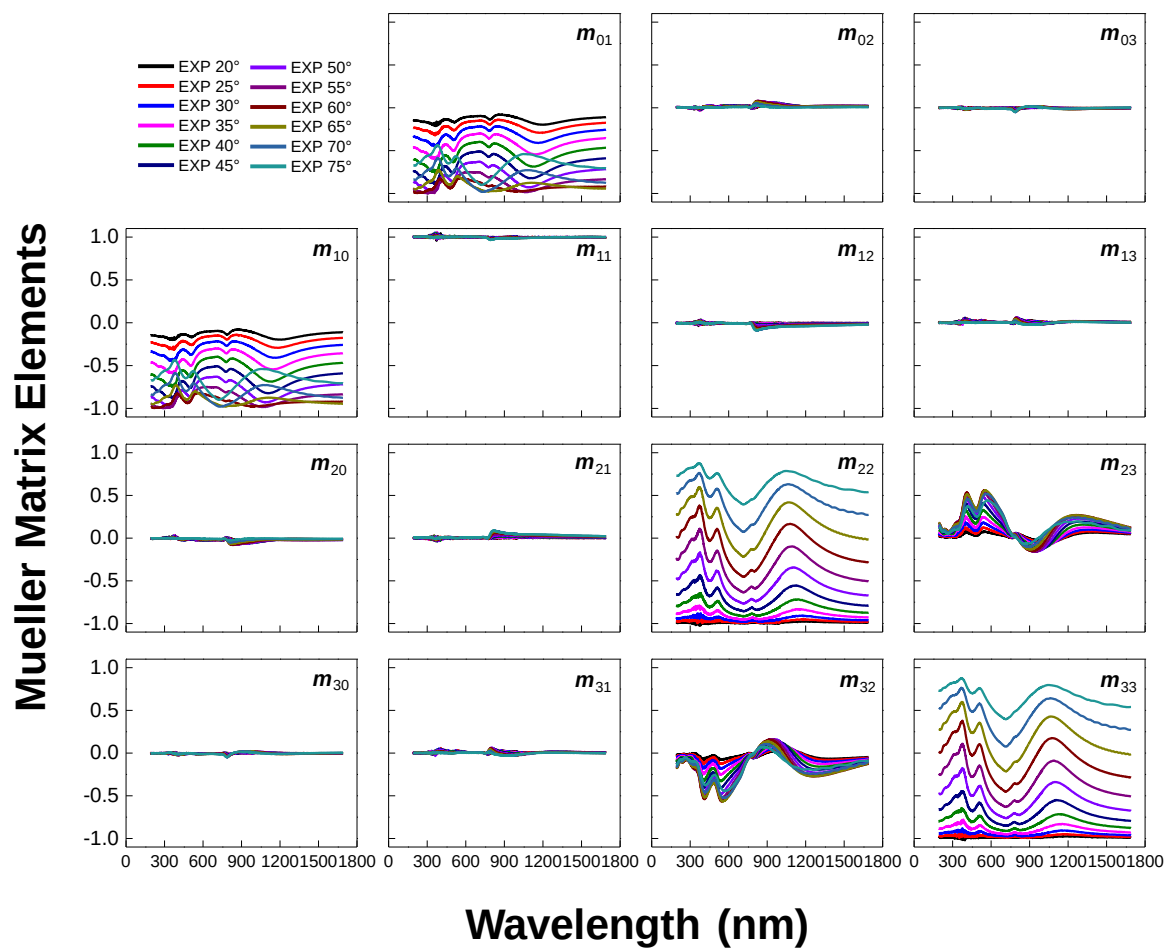
Supplementary Fig. 49 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in transmission mode with 90° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



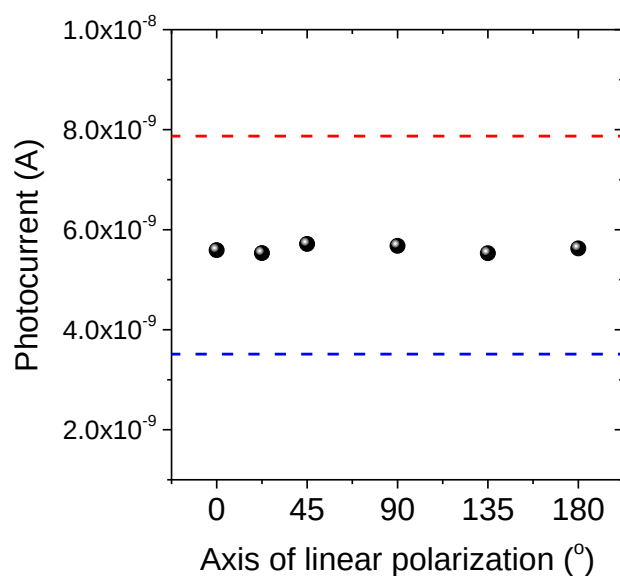
Supplementary Fig. 50 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in reflection mode with 0° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



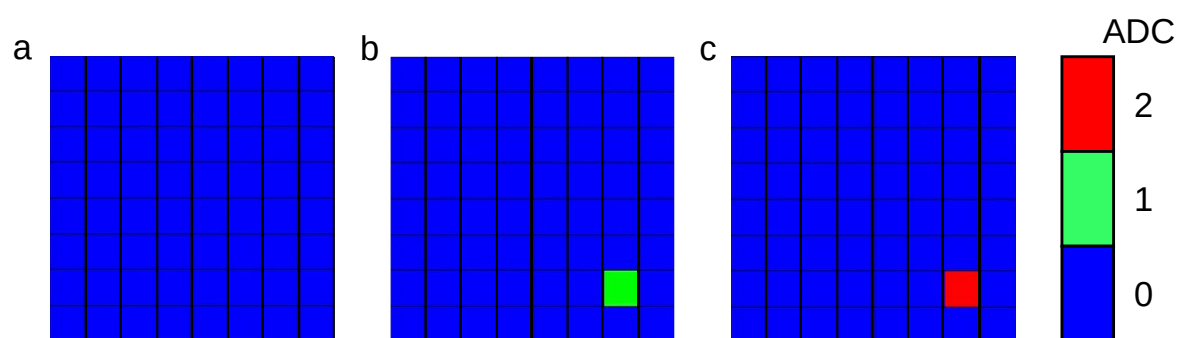
Supplementary Fig. 51 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in reflection mode with 45° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



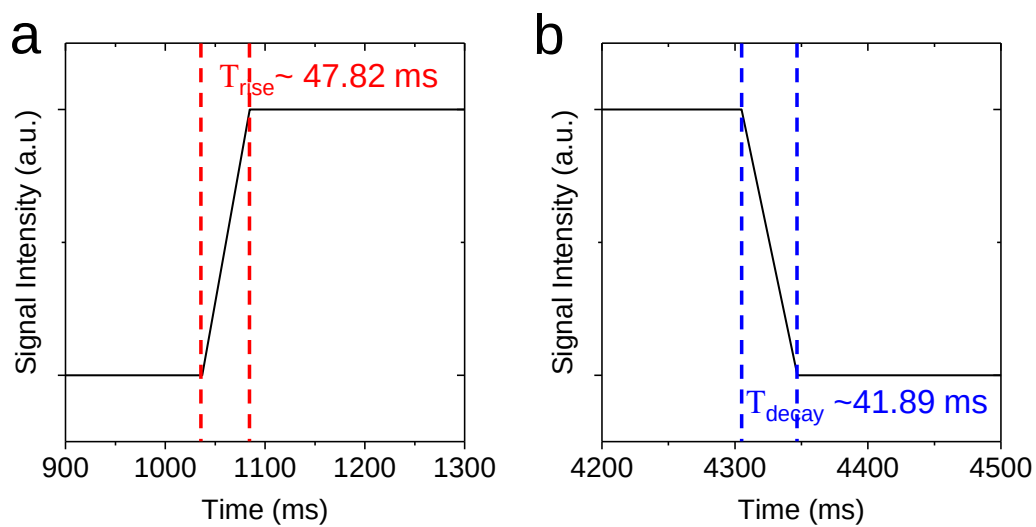
Supplementary Fig. 52 | MMSE for various contributions to optical activity for the S5011-doped PCPDTTBTT film. MMSE data were recorded in reflection mode with 90° azimuthal sample rotation. EXP indicates the installation angle between the incident light path and the normal light path to the substrate surface.



Supplementary Fig. 53 | Photocurrents of CP photodetectors under linearly polarized light. Photocurrents of CP photodetectors based on the S5011-doped PCPDTTBT films were recorded under the incidence of linearly polarized light with different linear polarization-to-sample axis orientations. Red and blue dotted lines indicate photocurrent levels under the RCP and LCP beams, respectively.



Supplementary Fig. 54 | Real-time spin helicity detection and imaging results. a–c, Polarization imaging results of the passive matrix array represented in customized software in the dark condition (**a**) and under LCP (**b**), and RCP (**c**) light irradiation.



Supplementary Fig. 55 | Time-dependent signal intensity of the passive matrix arrays. a,
b, Recorded signal intensity of the passive matrix arrays as a function of time with the circularly
irradiating polarized beam on **(a)** and off **(b)**.

Supplementary Table 1 | Surface energies of various surfaces calculated from the contact angle measurement.

Surface	Contact angle [deg]		γ_s^p ^a [mJ m ⁻²]	γ_s^d ^a [mJ m ⁻²]	γ_s ^b [mJ m ⁻²]
	Water	Glycerol			
Quartz	59.9	52.3	23.5	18.4	41.9
ITO	50.4	51.4	42.6	7.7	50.3
ZnO	49.0	63.3	70.5	0.0	70.5

^a γ_s^p and γ_s^d refer to the polar and dispersion components of the surface energy, respectively.

^b γ_s is the surface energy; $\gamma_s = \gamma_s^p + \gamma_s^d$.

Supplementary Table 2 | Summarized chiroptical and optoelectronic performances of electronic devices in reported studies.

Number (Ref)	Article	CD type	Operation	Voltage(V)	Wave length(nm)	$ g_{\text{abs}} $	$ g_{\text{ph}} $	EQE(%)
1 ^(S33)	<i>Adv. Mater.</i> 22 , E131–E134 (2010)	Organic	Photovoltaic	0	543	0.06	0.017	
2 ^(S34)	<i>Nat. Photon.</i> 7 , 634–638 (2013)	Organic	Phototransistor	$V_D = -60$ $V_G = -5$	365		1.8	0.1
3 ^(S35)	<i>Nat. Commun.</i> 6 , 8379 (2015)	Plasmonic	Photovoltaic	0	1340	1	1.1	0.2
4 ^(S36)	<i>Sci. Rep.</i> 6 , 19580 (2016)	Chiral mirror	Photoconductor	0.1	635	1.2	0.17	30
5 ^(S37)	<i>Adv. Mater.</i> 29 , 1605828 (2017)	Organic	Phototransistor	$V_D = 70$ $V_G = 70$	460	0.015	0.33	88100
6 ^(S38)	<i>Adv. Funct. Mater.</i> 29 , 1805279 (2019)	Chiral mirror	Phototransistor	$V_D = 1.5$ $V_G = -2$	405	1.6	0.67	
7 ^(S4)	<i>Adv. Funct. Mater.</i> 29 , 1808668 (2019)	Organic	Photoconductor	5	375		0.1	0.15
8 ^(S39)	<i>Adv. Funct. Mater.</i> 29 , 1900684 (2019)	Organic	Photovoltaic	0	543	0.09	0.1	50
9 ^(S40)	<i>Nat. Commun.</i> 10 , 1927 (2019)	Hybrid	Photoconductor	10	395	0.02	0.1	39
10 ^(S41)	<i>Opt. Lett.</i> 44 , 2998–3001 (2019)	Plasmonic	Photovoltaic	0	3800		1.3	1.1e-05
11 ^(S42)	<i>ACS Nano</i> 13 , 3659–3665 (2019)	Hybrid	Photoconductor	3	518		0.1	108
12 ^(S43)	<i>Angew. Chem. Int. Ed.</i> 59 , 6442– 6450 (2020)	Hybrid	Photoconductor	10	532	0.002	0.2	9.1
13 ^(S44)	<i>Sci. Adv.</i> 6 , 3274–3285 (2020)	Hybrid	Photoconductor	−0.5	395	0.04	1.85	87.5

14 ^(S45)	<i>J. Mater. Chem. C</i> 8 , 9271–9275 (2020)	Organic	Phototransistor	$V_D = -60$ $V_G = -60$	450		1.94	0.016
15 ^(S46)	<i>ACS Nano</i> 14 , 14146–14156 (2020)	Organic	Phototransistor	$V_D = 10$ $V_G = 10$	495		0.129	3e5
16 ^(S32)	<i>Adv. Funct. Mater.</i> 30 , 2006236 (2020)	Chiral mirror	Phototransistor	$V_D = -60$ $V_G = -3$	830		1.9	4.5e4
17 ^(S47)	<i>Adv. Mater.</i> 33 , 2004115 (2021)	Organic	Phototransistor	$V_D = 20$ $V_G = 10$	405	0.0006	1.27	
18 ^(S2)	<i>Adv. Opt. Mater.</i> 10 , 2101044 (2022)	Organic	Photoconductor	-0.5 -1	510 455	0.03 0.2	0.41 0.12	8.4 5.2
19 ^(S48)	<i>Nat. Commun.</i> 12 , 142 (2021)	Organic	Phototransistor	$V_D = -80$ $V_G = -40$	730	0.002	0.057	89
20 ^(S49)	<i>ACS Nano</i> 15 , 7608–7617 (2021)	Hybrid	Phototransistor	$V_D = -2$ $V_G = 0$	405	0.056	0.25	1.38e5
21 ^(S50)	<i>Angew. Chem. Int. Ed.</i> 60 , 8415–8418 (2021)	Hybrid	Photovoltaic Photoconductor	0 5	520	0.0015	0.3 0.22	0.005
22 ^(S51)	<i>ACS Mater. Lett.</i> 4 , 401–409 (2022)	Organic	Photovoltaic	0	606	0.02	0.17	57
23 ^(S52)	<i>Adv. Sci.</i> 9 , 2104598 (2022)	Plasmonic	Photovoltaic	0	808	0.02	0.47	77.7
24	(This work)	Organic	Photoconductor	-10	405	0.84	1.02	120
					670	1.21	1.16	11

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