Electronic Supplementary Information

Chirality-Activated Mechanoluminescence from Aggregation-

Induced Emission Enantiomers with High Contrast

Mechanochromism and Force-Induced Delayed Fluorescence

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1. Experimental Procedures

Chemical Reagents and Materials.

4-Bromophthalic anhydride, tris(dibenzylideneacetone)dipalladium, (S)-(+)-1,2,3,4-tetrahydro-1naphthylamine (*ee*>98%), (*R*)-(-)-1,2,3,4-tetrahydro-1-naphthylamine (*ee*>98%), phenothiazine, dicyclohexyl(2',6'-diisopropoxy-[1,1'-biphenyl]-2-yl)phosphine, potassium phosphate tribasic and super dry *N*,*N*-dimethylformamide were purchased from Aladdin and used as received. All other solvents and reagents were purchased from Guangzhou Zeyuan Company (China) with analytical grade and used without further purification.

Measurements

Nuclear magnetic resonance, including ¹H NMR and ¹³C NMR, were conducted on a Bruker AVANCE 600 MHz spectrometer by using tetramethylsilane as internal standard and CDCl₃ as solvent. Elemental analysis was carried out on a Vario EL analyzer. Mass spectra were measured on DSQ & MAT95XP-HRMS thermospectrometers. Powder XRD patterns were recorded by a Rigaku X-ray diffractometer (Ultima IV) in a scanning rate of 4° (2 θ)/min at 293 K. The thermal behaviors of the samples were determined by differential scanning calorimetry (DSC) at a heating rate of 10 °C/min under N₂ atmosphere using a TA thermal analyzer (Q20). PL and ML spectra were collected on an Ocean Optics spectrophotometer (QE65 Pro). Time-resolved emission decay curves and absolute PL quantum yields (PLQYs) were obtained by using a spectrometer (FLS980) equipped with a calibrated integrating sphere from Edinburgh Instruments. Circular dichroism (CD) spectra were characterized on a CD spectrometer (J-815) from JASCO instrument. Circularly polarized luminescence (CPL) spectra and the corresponding dissymmetry factors were measured on a commercialized instrument JASCO CPL-300 spectrophotometer at a scanning rate of 50 nm/min at room temperature. Data of single-crystal X-ray diffraction was achieved from an Agilent Technologies Gemini A Ultra system with Cu-K α radiation ($\lambda = 1.54178$ Å) at 150 K. The crystal structures were solved by using direct methods following the difference Fourier syntheses. All nonhydrogen atoms of the molecules were refined anisotropically through least-squares on F^2 by utilizing the Olex2 program suite. CCDC 1906044, 1906045 and 1906046 contain the crystallographic data for this paper. Photoluminescent images and videos recording the mechanoluminescence phenomena were taken by a digital camera (Canon T5i) with a Canon 50 mm f/1.8G STM camera lens. ML images of the samples were picked out from the videos.

Theoretical calculations

Theoretical calculations based on DFT and TD-DFT were conducted on a Gaussian 16 program.¹ The molecular geometries of (*R*)-ImNT and (*S*)-ImNT were directly selected from single crystal structures and were submitted for simulation without further optimization to evaluate their dipole moments. Stable conformations of (*R*)-ImNT and (*S*)-ImNT at the lowest energy states were searched by using DFT and M062X at the 6–311G** level. Subsequently, exciton energies for the most possible quasi-equatorial conformations of the enantiomers in singlet (S₁) and triplet states (T₁) were estimated through a combination of TD-DFT and M062X at the 6–311G** level. Kohn-Sham

frontier orbital analysis was performed based on the results of theoretical calculation to elucidate the changes of intramolecular charge transfer strength for the molecules in pristine and ground samples. Intermolecular interaction analyses for the molecules in single crystal structures were carried out by Multiwfn with independent gradient model (IGM),² and the results were plotted via VMD software (version 1.9.3).³

Synthesis



Scheme S1. Synthetic routes of the target compounds.

General procedure for the synthesis of chiral luminophores with Mechanoluminescence

(R)-5-bromo-2-(1,2,3,4-tetrahydronaphthalen-1-yl)isoindoline-1,3-dione [(R)-ImBr] To a solution of 4-bromophthalic anhydride (1.50 g, 6.60 mmol) in super dry N,N-dimethylformamide (15 ml) (R)-(-)-1,2,3,4-tetrahydro-1-naphthylamine (0.97 g, 6.60 mmol) was added. The mixture was stirred at 150 °C under argon for 24 h. After cooling to room temperature, the mixture was poured to 150 mL saturated NaCl solution, and the precipitate was filtered and washed with water for several times. The crude product was purified by silica gel column chromatography with dichloromethane/petroleum (v/v=1:2) as eluent to give (R)-ImBr as a white powder (1.82 g, Yield 78%). ¹H NMR (600 MHz, CDCl₃) δ 7.98 – 7.94 (s, 1H), 7.87 – 7.84 (dd, J = 7.9, 1.5 Hz, 1H), 7.71 -7.68 (d, J = 7.9, 1H), 7.15 - 7.13 (d, J = 4.2, 2H), 7.07 - 7.03 (m, 1H), 6.92 - 6.89 (d, J = 7.7 Hz, 1H), 5.54 - 5.51 (dd, J = 10.8, 5.8 Hz, 1H), 3.05 - 2.98 (m, 1H), 2.85 - 2.80 (m, 1H), 2.42 - 2.35 (m, 1H), 2.13 - 2.07 (m, 2H), 1.88 - 1.81 (m, 1H). ¹³C NMR (151 MHz, CDCl₃) δ 167.30, 166.72, 137.87, 136.97, 134.28, 133.72, 130.59, 129.31, 128.90, 127.05, 126.68, 126.13, 125.75, 124.68, 53.43, 49.57, 29.34, 28.04, 22.32. HRMS m/z: [M]⁺ calcd for C₁₈H₁₄BrNO₂, 355.0208; found, 355.0201. Anal. calcd for C₁₈H₁₄BrNO₂ (%): C 60.69, H 3.96, N 3.93; found: C 60.60, H 3.98, N 3.95.

(*S*)-5-bromo-2-(1,2,3,4-tetrahydronaphthalen-1-yl)isoindoline-1,3-dione **[(S)-ImBr]** Following the same synthetic approach for (*R*)-ImBr, the reaction of 4-bromophthalic anhydride (1.50 g, 6.60 mmol) and (*S*)-(-)-1,2,3,4-tetrahydro-1-naphthylamine (0.97 g, 6.60 mmol) was conducted to produce (*S*)-ImBr as a white powder (1.88 g, Yield 80%). ¹H NMR (600 MHz, CDCl₃) δ 7.98 – 7.94 (s, 1H), 7.87 – 7.84 (dd, *J* = 7.9, 1.5 Hz, 1H), 7.71 – 7.68 (d, *J* = 7.9, 1H), 7.15 – 7.13 (d, *J* = 4.2, 2H), 7.07 – 7.03 (m, 1H), 6.92 – 6.89 (d, *J* = 7.7 Hz, 1H), 5.54 – 5.51 (dd, *J* = 10.8, 5.8 Hz, 1H), 3.05 – 2.98 (m, 1H), 2.85 – 2.80 (m, 1H), 2.42 – 2.35 (m, 1H), 2.13 – 2.07 (m, 2H), 1.88 – 1.81 (m, 1H). ¹³C NMR (151 MHz, CDCl₃) δ 167.29, 166.72, 137.87, 136.97, 134.27, 133.72, 130.59, 129.31, 128.90, 127.04, 126.67, 126.13, 125.75, 124.67, 49.57, 29.34, 28.03, 22.32. HRMS m/z: [M]⁺ calcd for C₁₈H₁₄BrNO₂, 355.0208; found, 355.0200. Anal. calcd for C₁₈H₁₄BrNO₂ (%): C 60.69, H 3.96, N 3.93; found: C 60.58, H 3.98, N 3.96.

(R)-5-(10H-phenothiazin-10-yl)-2-(1,2,3,4-tetrahydronaphthalen-1-yl) isoindoline-1,3-dione [(R)-**ImNT**] A flask was charged with (*R*)-ImBr (1.78 g, 5.00 mmol), phenothiazine (1.49 g, 7.50 mmol), Pd₂(dba)₃ (0.05 g, 0.087 mmol), Ruphos (0.15 g, 0.32 mmol), K₃PO₄ (4.24 g, 20.00 mmol) and toluene (20 mL). The mixture was then degassed and stirred at 120 °C under argon for 24 h. After cooling to room temperature, the reaction solution was concentrated, and the resulting crude product was purified by silica gel column chromatography with dichloromethane/petroleum (v/v=2:3) as eluent. (R)-ImNT was further recrystallized from dichloromethane/methanol to give a light green powder in 71% yield. (1.68 g). ¹H NMR (600 MHz, CDCl₃) δ 7.64 – 7.61 (d, J = 8.4 Hz, 1H), 7.48 -7.45 (d, J = 7.7 Hz, 2H), 7.40 - 7.38 (s, 1H), 7.37 - 7.33 (m, 4H), 7.26 - 7.21 (dt, J = 14.4, 4.0Hz, 3H), 7.13 – 7.10 (d, J = 4.1 Hz, 2H), 7.05 – 7.02 (m, 1H), 6.93 – 6.91 (d, J = 7.7 Hz, 1H), 5.49 -5.45 (m, 1H), 3.02 - 2.96 (m, 1H), 2.83 - 2.78 (m, 1H), 2.38 - 2.32 (m, 1H), 2.10 - 2.03 (m, 2H), 1.86 - 1.79 (m, 1H). ¹³C NMR (151 MHz, CDCl₃) δ 168.06, 167.91, 151.12, 140.95, 137.84, 134.83, 134.29, 133.66, 129.14, 129.01, 127.60, 126.81, 126.50, 126.39, 126.02, 125.86, 124.76, 123.74, 119.92, 110.54, 49.12, 29.38, 28.07, 22.35. EI-MS m/z: [M]⁺ calcd for C₃₀H₂₂N₂O₂S, 474; found, 474. HRMS m/z: [M]⁺ calcd for C₃₀H₂₂N₂O₂S, 474.1402; found, 474.1392. Anal. calcd for C₃₀H₂₂N₂O₂S (%): C 75.93, H 4.67, N 5.90, S 6.76; found: C 75.80, H 4.69, N 5.93, S 6.72. Single crystals for X-ray analysis were isolated by the slow solvent evaporation of a solution of (R)-ImNT in a mixture of methanol/dichloromethane.

(*S*)-5-(10H-phenothiazin-10-yl)-2-(1,2,3,4-tetrahydronaphthalen-1-yl)isoindoline-1,3-dione [(*S*)-ImNT] Following the same synthetic approach for (*R*)-ImNT, the reaction of (*S*)-ImBr (1.78 g, 5.00 mmol), phenothiazine (1.49 g, 7.50 mmol), Pd₂(dba)₃ (0.05 g, 0.087 mmol), Ruphos (0.15 g, 0.32 mmol) and K₃PO₄ (4.24 g, 20.00 mmol) was conducted to produce (*S*)-ImNT as a light green powder (1.88 g, Yield 79%). ¹H NMR (600 MHz, CDCl₃) δ 7.65 – 7.60 (d, *J* = 8.3 Hz, 1H), 7.49 – 7.45 (d, *J* = 7.8 Hz, 2H), 7.40 – 7.38 (s, 1H), 7.37 – 7.32 (m, 4H), 7.26 – 7.21 (dt, *J* = 14.4, 4.0 Hz, 3H), 7.13 – 7.10 (d, *J* = 4.0 Hz, 2H), 7.06 – 7.02 (m, 1H), 6.94 – 6.90 (d, *J* = 7.7 Hz, 1H), 5.49 – 5.44 (m, 1H), 3.02 – 2.96 (m, 1H), 2.83 – 2.78 (m, 1H), 2.38 – 2.32 (m, 1H), 2.11 – 2.03 (m, 2H), 1.87 – 1.79 (m, 1H). ¹³C NMR (151 MHz, CDCl₃) δ 168.06, 167.91, 151.12, 140.95, 137.84, 134.83, 134.29, 133.66, 129.14, 129.02, 127.60, 126.81, 126.50, 126.39, 126.02, 125.86, 124.76, 123.74, 119.92, 110.54, 49.12, 29.38, 28.07, 22.35. EI-MS m/z: [M]⁺ calcd for C₃₀H₂₂N₂O₂S, 474; found, 474. HRMS m/z: [M]⁺ calcd for C₃₀H₂₂N₂O₂S (%): C 75.93, H 4.67, N 5.90, S 6.76; found: C 75.83, H 4.70, N 5.92, S 6.73. Single

crystals for X-ray analysis were isolated by the slow solvent evaporation of a solution of (S)-ImNT in a mixture of methanol/dichloromethane.

Preparation of (R, S)-ImNT Large amounts of methanol was added into a concentrated dichloromethane solution that contains (R)-ImNT and (S)-ImNT with molar ratio of 1:1 under the action of ultrasound. Subsequently, the resulting precipitate was filtered and dried to give (R, S)-ImNT as a light green powder. Single crystals for X-ray analysis were isolated by the slow solvent evaporation of a solution of (R, S)-ImNT in a mixture of methanol/dichloromethane.

2. Results and Disscussion



I. AIE Properties

Fig. S1 PL spectra of the dilute solutions of (R, S)-ImNT a) and (R)-ImNT b) in water/THF mixtures with different water fractions (f_w) . (Concentration: 0.1 mM).



Fig. S2 UV-visible absorption spectra of the dilute solutions of (R, S)-ImNT a), (R)-ImNT b) and (S)-ImNT c) in water/THF mixtures with different water fractions (f_w) . (Concentration: 0.1 mM).

II. Photophysical and Morphological Properties



Fig. S3 Emission decay curves of the pristine samples of enantiomers and racemate at ambient condition.



Fig. S4 Circular dichroism spectra of the dilute solutions of (R)-ImNT, (S)-ImNT and (R,S)-ImNT in dichloromethane. (Concentration: 0.1 mM).

Circular dichroism (CD) spectra of the enantiomers and the racemate were measured in dilute solutions to investigate their chiroptical properties in the ground states. As depicted in Fig. S6, (*R*)-ImNT and (*S*)-ImNT show mirror-image bands in the CD spectra. The evident positive and negative Cotton effects of the enantiomers at around 285 nm and 323 nm correspond to the π - π * transition absorptions of the molecules. Meanwhile, there are also weak Cotton effects at around 377 nm, which can be assigned to the intramolecular charge transfer (ICT) transitions from phenothiazine to phthalimide. As evidenced by these important signals, the chirality of tetrahydronaphthalene unit is successfully transmitted to the achiral phenothiazine and phthalimide moieties. In contrast, no CD signal can be recorded for the racemate, indicating its achiral nature.

CPL measurements for (R)-ImNT, (S)-ImNT and (R,S)-ImNT in dilute solutions were also implemented to study their chiroptical properties in the excited states. However, signals of the enantiomers and the racemate were both too weak to be recorded, which may be ascribed to the prominent AIE properties and the achiral characteristic, respectively.



Fig. S5 Dissymmetric factors (g_{lum}) of the as-prepared samples of (R)-ImNT, (S)-ImNT and (R,S)-ImNT.



Fig. S6 a) DSC curves of the as-prepared samples of (R,S)-ImNT, (R)-ImNT and (S)-ImNT. b) Experimental XRD patterns of the crystalline powders and simulated XRD patterns of the single crystals of (R,S)-ImNT, (R)-ImNT and (S)-ImNT.

III. Results of Single Crystal Analysis and Multiwfn Simulation

Compound	Crystal System	Space Group	Symmetry	Polarity	ML Activity
(<i>R,S</i>)-ImNT	Monoclinic	$P2_{1}/c$	Centrosymmetric (achiral)	Non-polar	Inactive
(S)-ImNT	Triclinic	<i>P</i> 1	Chiral	Polar	Active
(R)-ImNT	Triclinic	<i>P</i> 1	Chiral	Polar	Active

Table S1. Photophysical properties and single crystal parameters of the compounds



Fig. S7 a) Molecular conformations of (*R*)-ImNT and (*S*)-ImNT molecules in the single crystal structure of (*R*, *S*)-ImNT. b) Molecular packing mode of (*R*)-ImNT and (*S*)-ImNT molecules in the unit cell of (*R*, *S*)-ImNT. D.M. means net dipolar moment of the molecules in unit cell. c) Distribution and strength of intramolecular interactions in the unit cell of (*R*, *S*)-ImNT.



Fig. S8 Molecular packing modes in the single crystal structures of (R, S)-ImNT a), (S)-ImNT b) and (R)-ImNT c).



Fig. S9 Intermolecular interactions in the single crystal structures of (*S*)-ImNT a), (*R*)-ImNT b) and (*R*, *S*)-ImNT c).

For (S)-ImNT, seven types of C-H···O interactions (with distances of 2.360, 2.487, 2.494, 2.516,

2.588, 2.639 and 2.719 Å), twelve types of C-H··· π interactions (with distances of 2.366, 2.678, 2.723, 2.761, 2.762, 2.788, 2.808, 2.818, 2.823, 2.825, 2.885 and 2.894 Å) and one type of C-H···S interaction (with distance of 2.995 Å) were observed in the single crystal structure. Similarly, seven types of C-H···O interactions (with distances of 2.357, 2.495, 2.497, 2.516, 2.586, 2.644 and 2.716, Å), eleven types of C-H··· π interactions (with distances of 2.673, 2.724, 2.765, 2.766, 2.788, 2.807, 2.815, 2.825, 2.828, 2.883 and 2.893 Å) and one type of C-H···S interaction (with distance of 2.995 Å) were found in the single crystal structure of (*R*)-ImNT. These intense multiple interactions help to solidify the molecular conformations and block the non-radiative pathways via restricting the intramolecular motions of the molecules. However, in the case of (*R*, *S*)-ImNT, only four C-H···O interactions (one type with distance of 2.503 Å) and eight C-H··· π interactions (two types with distances of 2.783 and 2.835 Å) were formed in single crystal structure, indicating a relatively loose molecular packing mode.



Fig. S10 a) Molecular conformations of (R)-ImNT molecules in single crystal structure. b) Molecular packing mode of (R)-ImNT molecules in the unit cell. D.M. means net dipolar moment of the molecules in unit cell. c) Distribution and strength of intramolecular interactions in the unit cell of (R)-ImNT.

IV. Alterations of Emission Properties Triggered by Force



Fig. S11 a) Emission decay curve of the well-ground sample of (*S*)-ImNT at ambient condition. b) PL spectra of the well-ground sample of (*S*)-ImNT under different conditions. c) Emission switching of (*S*)-ImNT under the stimuli of mechanical force and dichloromethane (DCM) vapor. After grinding, the emission maxima can be recovered by fuming with DCM vapor for about 15 min.



Fig. S12 Variations in emission properties of (*R*)-ImNT triggered by external force. a) Changes of PL spectra of (*R*)-ImNT under grinding. "×0.2" and "×0.4" mean that intensities of the spectra presented in the figure are one fifth and two fifths of those of the original ones, respectively. The inset depicts that the CIE_{x,y} coordinates of (*R*)-ImNT linearly change from (0.16, 0.29) to (0.54, 0.43) under grinding. b) Emission decay curve of the well-ground sample at ambient condition.



Fig. S13 Variations in emission properties of (R, S)-ImNT triggered by external force. a) Changes of PL spectra of (R, S)-ImNT under grinding. "×0.2" and "×0.5" mean that intensities of the spectra presented in the figure are one fifth and a half of those of the original ones, respectively. The inset depicts that the CIE_{x,y} coordinates of (R, S)-ImNT linearly change from (0.19, 0.31) to (0.54, 0.43) under grinding. b) Emission decay curve of the well-ground sample at ambient condition.



Fig. S14 PL spectra of the well-ground sample of (R)-ImNT a) and (R, S)-ImNT b) under different conditions.

Emission spectra in the absence and presence of oxygen for the well-ground samples of enantiomers and racemate were shown in figures S11b and S14. The ratio of prompt and delayed fluorescence is given by $I_{02}/(I_{no,02}-I_{02})$, where I_{02} and $I_{no 02}$ are the integral of emission with and without oxygen, respectively.⁴ Obviously, luminescence of the red-light emitting powders was partially quenched in the present of oxygen, further verifying their TADF characteristics. The ratios of prompt and delayed fluorescence for the samples of (*S*)-ImNT, (*R*)-ImNT and (*R*, *S*)-ImNT are determined to be 9.12:1, 9.42:1 and 4.95:1, respectively. That means, the proportions of delayed fluroescence are 9.88%, 9.59% and 16.81% to the total luminescence under vacuum.



Fig. S15 Emission decay curves of the well-ground samples of (R)-ImNT a) and (R, S)-ImNT b) at different temperatures under vacuum.



Fig. S16 Emission switching of (R)-ImNT a) and (R, S)-ImNT b) under the stimuli of mechanical force and DCM vapor. After grinding, the emission maxima can be recovered by fuming with DCM vapor for about 15 min.

V. Mechanism for the Variations in Emission Properties



Fig. S17 Changes of XRD patterns for the samples of (*R*)-ImNT a), (*S*)-ImNT b) and (*R*, *S*)-ImNT c) upon grinding.



Fig. S18 Changes of DSC curves for the samples of (*R*)-ImNT a), (*S*)-ImNT b) and (*R*, *S*)-ImNT c) upon grinding.



Fig. S19 Kohn-Sham frontier orbitals of the molecules with different conformations in the single crystal structures of (R)-ImNT a), (S)-ImNT b) and (R, S)-ImNT c), respectively.



Fig. S20 Fluorescence and phosphorescence spectra of pristine samples of (*R*)-ImNT a), (*S*)-ImNT b) and (*R*, *S*)-ImNT c). Their ΔE_{ST} values were obtained from the onsets of fluorescence and phosphorescence spectral profiles.



Fig. S21 a) Stable conformations of (*S*)-ImNT obtained from theoretical calculation. b) Stable conformations of (*R*)-ImNT obtained from theoretical calculation. c) Kohn-Sham frontier orbitals of (*R*)-ImNT in the most possible quasi-equatorial conformation.



Fig. S22 Fluorescence and phosphorescence spectra of the well-ground samples of (*R*)-ImNT a), (*S*)-ImNT b) and (*R*, *S*)-ImNT c). Their ΔE_{ST} values were obtained from the onsets of fluorescence and phosphorescence spectral profiles.

VI. ¹H NMR, ¹³C NMR, Mass Spectra and Single Crystal Structural Information of the compounds



Fig. S23 ¹H NMR spectrum of (*R*)-ImBr (In CDCl₃).







Fig. S26 ¹H NMR spectrum of (*S*)-ImBr (In CDCl₃).







Fig. S28 HRMS of (S)-ImBr.



Fig. S30 13 C NMR spectrum of (*R*)-ImNT (In CDCl₃).







Fig. S32 HRMS of (*R*)-ImNT.



Fig. S34 ¹³C NMR spectrum of (S)-ImNT (In CDCl₃).







Fig. S36 HRMS of (*S*)-ImNT.

Compound	(R)-ImNT	(S)-ImNT	(<i>R</i> , <i>S</i>)-ImNT
formula	$C_{30}H_{22}N_2O_2S$	$C_{30}H_{22}N_2O_2S$	$C_{30}H_{22}N_2O_2S$
fw	474.1402	474.1402	474.1402
crystal system,	triclinic	triclinic	monoclinic
T/\mathbf{K}	150.00(10)	150.00(10)	150.00(10)
space group	<i>P</i> 1	<i>P</i> 1	$P2_1/c$
a/Å	12.1382(2)	12.1350(2)	14.4773(2)
b /Å	12.8179(3)	12.8327(2)	12.17882(16)
c/Å	15.1854(3)	15.1864(2)	13.40691(18)
lpha/ °	85.684(2)	85.7850(10)	90
<i>β</i> / °	78.373(2)	78.3710(10)	104.9253(14)
γ/°	89.276(2)	89.3450(10)	90
$V/\text{\AA}^3, Z$	2307.59(8), 4	2310.08(6), 4	2284.11(6), 4
F (000)	992	992	992
crystal size / mm ³	0.18×0.16×0.10	0.40×0.20×0.10	0.25×0.17×0.15
reflections collected /	30459/14132	37878/17289	13740/4549
unique (R_{int})	$(R_{\rm int} = 0.0331)$	$(R_{\rm int} = 0.0275)$	$(R_{\rm int} = 0.0314)$
obsd reflns [$I \ge 2\sigma(I)$]	13566	17091	4275
data/restraints/parameter	14132/3/1261	17289/3/1261	4549/0/316
$D_c/\mathrm{Mg}\cdot\mathrm{m}^{-3}$	1.366	1.364	1.380
μ / mm ⁻¹	1.497	1.495	1.512
goodness-of-fit on F^2	1.033	1.030	1.072
$R_1, {}^{[a]} w R_2 {}^{[b]} [I \ge 2\sigma(I)]$	0.0494, 0.1369	0.0290, 0.0783	0.0422, 0.1103
R_1 , wR_2 (all data)	0.0537, 0.1442	0.0293, 0.0789	0.0444, 0.1124

Table S2. Crystal data and structure refinement for the three single crystals

^a $R_I = \Sigma ||F_o| - |F_c|| \Sigma |F_o|$. ^b $wR_2 = [\Sigma[w(F_o^2 - F_c^2)^2] \Sigma w(F_o^2)^2]^{1/2}$, where $w = 1/[^2(F_o)^2 + (aP)^2 + bP]$ and $P = (F_o^2 + 2F_c^2)/3$

3. Reference

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4. Figures for the Article



Fig. 1 Chemical structures of (R)-ImNT and (S)-ImNT.



Fig. 2 a) PL spectra of the dilute solutions of (*S*)-ImNT in water/THF mixtures with different water fractions (f_w). (Concentration: 0.1 mM). The insets depict the emission images of (*S*)-ImNT in pure THF and in water/THF mixture with 90% water fraction under the illumination of 365 nm UV light. b) PL spectra of the compounds in the solid state at ambient condition.



Fig. 3 CPL a) and ML spectra b) of the as-prepared samples of (R,S)-ImNT, (R)-ImNT and (S)-ImNT at ambient condition. The insets in b) depict the ML images of (R)-ImNT and (S)-ImNT in daylight.



Fig. 4 a) Molecular conformations of (*S*)-ImNT molecules in single crystal structure. The digits in the figure represent bent angle, dihedral angle or dipolar moment. Take conformation S1 as an example. Its dipolar moment is 4.41 debye. While the bent angle between the phenothiazine moiety and its neighboring phenyl ring is 115.89°, the dihedral angle between plane A and plane B is 66.02° . b) Molecular packing mode of (*S*)-ImNT molecules in the unit cell. D.M. means net dipolar moment of the molecules in unit cell. c) Distribution and strength of intramolecular interactions in the unit cell of (*S*)-ImNT. d) Plots of the IGM with isovalue of 0.005 versus the electron density.



Fig. 5 Variations in emission properties of (*S*)-ImNT triggered by external force. a) Changes of PL spectra of (*S*)-ImNT under grinding. "×0.2" and "×0.4" mean that intensities of the spectra presented in the figure are one fifth and two fifths of those of the original ones, respectively. The inset depicts linear changes of $CIE_{x,y}$ coordinates of (*S*)-ImNT under grinding. b) Luminescent images of the samples upon grinding treatments (under the illumination of 365 nm UV light). c) Emission decay curves of the well-ground sample at different temperatures under vacuum. The insets are the Kohn-Sham frontier orbitals of (*S*)-ImNT in the most possible quasi-equatorial conformation.