Electronic Supplementary Information for

Solvent-Regulated Chiral Exciton Coupling and CPL Sign Inversion of an Amphiphilic Glutamide-Cyanostilbene

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1. Synthetic procedures to GCS.

Compound GluCN was reported by our group.¹ In general, *L*-GluCN or *D*-GluCN (0.67 g, 1.0 mmol) and triethylamine (0.10 g, 1.0 mmol) were dissolved in 15 mL N,N-dimethylformamide (DMF) and stirred for 30 minutes, 1-Hydroxybenzotriazole (HOBt) (0.151 g, 1.1 mmol), 1-(3-Dimethylaminopropyl)-3-ethylcarbodiimide hydrochloride (EDC-HCl) (0.22 g, 1.1 mmol) and stearic acid (0.28 g, 1.0 mmol) were added to the solution and stirred for 72 hours at room temperature. White precipitate was obtained after filtration by Buchner funnel, and the samples were dried by vacuum drying chamber to remove the residual solvent. White solid (0.45 g, 0.55 mmol), yield 55.1 %.



Scheme S1 Synthetic routes to GCS.

¹H NMR for *L*-**GCS** and *D*-**GCS** (400 MHz, d₆-DMSO, δ , ppm): 0.82-0.85 (t, 3H, CH₃), 1.18 (m, 28H, CH₂), 1.47-1.51 (m, 2H, CH₂), 1.92-2.01 (m, 1H, CH₂), 2.03-2.12 (m, 1H, CH₂), 2.14-2.17 (t, 2H, CH₂), 2.43-2.47 (m, 2H, CH₂), 4.45-4.51 (m, 1H, CH), 7.46-7.55 (m, 6H, Ar-H), 7.68-7.78 (m, 8H, Ar-H), 7.89-7.92 (m, 4H, Ar-H), 7.95 (s, 1H, CH), 7.97 (s, 1H, CH), 8.13-8.15 (d, 1H, NH), 10.18 (s, 1H, NH), 10.30 (s, 1H, NH). ¹³C NMR for **GCS** (400 MHz, d₆-DMSO, δ , ppm): 15.76, 15.79, 23.92, 23.95, 27.11, 30.49, 30.54, 30.57, 30.68, 30.85, 30.88, 30.90, 30.93, 30.96, 33.13, 33.15, 37.01, 54.94, 111.93, 111.98, 119.72, 121.10, 121.15, 121.48, 128.11, 128.15, 130.76, 130.81, 130.84, 141.75, 142.12,142.82, 143.04, 172.48, 172.63, 174.40. MALDI-TOF-MS (m/z): [M + Na]⁺ calculated for C₅₃H₆₃N₅O₃Na⁺

, 840.48; found, 840.70. Elemental analysis: calculated for C, 77.81; H, 7.76; N, 8.56; found C, 77.50; H, 7.73; N, 8.42.

2. Supplementary Figures.



Fig. S1 The assignment of hydrogen atoms and 2D-COSY NMR spectrum of *L*-GCS, (400 MHz, d_6 -DMSO, δ , ppm, 6.11 mM).



Fig. S2 2D-NOESY NMR spectrum of *L*-GCS, (400 MHz, d_6 -DMSO, δ , ppm, 6.11 mM). The NOE effect between hydrogens j and c, d was highlighted in the DFT-optimized structure.



Fig. S3 Molecular orbitals and energy levels for *L*-**GCS**. Gaussian 09, B3LYP 6-311 G(d,p) level.² The blue and green colors of electron cloud represent hole and electron, respectively. The red, cyan orange, blue and gray balls represent oxygen, carbon, nitrogen, and hydrogen atoms, respectively.³

Table S1 Excited states and MO transition contributions ^a of L-GCS

States	Main MO contributions	Energy / eV	Wavelength / nm	Oscillator strength
S1	HOMO \rightarrow LUMO (99.6%)	3.21	386	0.0001
S2	HOMO \rightarrow LUMO+1 (84.4%)	3.38	367	1.0705
S3	HOMO-1 \rightarrow LUMO (84.5%)	3.49	355	0.7889

^a Calculated by TD-DFT, Gaussian 09, B3LYP 6-311 G(d,p) level.



Fig. S4 (a) Normalized UV-vis absorption and fluorescence spectra of *L*-GCS gel in CCl_4 . (b) CD spectra of *L*-GCS (black line) and *D*-GCS (red line) gels in CCl_4 , [GCS]=6.11 mM.

 Table S2 Summary of optical properties of GCS in different solvents.

Solvents	Absorption maxima (nm)	Emission maxima (nm)	Stokes shift (nm)	Quantum yields
DMF	341	433	92	1.2 %
$DMF/H_2O, f_w = 40 \%$	358	437	79	1.5 %
Toluene	329	424	95	0.6 %



Fig. S5 CD spectra of GCS in dilute (a) toluene and (b) CCl₄. The black lines are corresponding to *L*-GCS and red lines are *D*-GCS. [GCS] = 1.0×10^{-5} M, cuvettes path length was 1 cm.



Fig. S6 Schematic illustration of intramolecular chiral exciton couplings of *L*-**GCS** and *D*-**GCS** in monomeric state.



Fig. S7 CD spectra of *L*-**GCS** (black line) and *D*-**GCS** (red line) gels in (a) DMSO and (b) MeCN, [**GCS**]=6.11 mM. The inserted figures are the inverted vials after heating-cooling protocols.



Fig. S8 CD spectra of *L*-GCS (black line) and *D*-GCS (red line) in DMF/H₂O cosolvents at (a) $f_w = 10$ % (b) $f_w = 20$ %, (c) $f_w = 30$ %, (d) $f_w = 50$ %, [GCS]=6.11 mM.



Fig. S9 CD spectrum of the GCS in DMSO/H₂O co-solvents, (a) $f_w = 10$ %, (b) $f_w = 20$ %, (c) $f_w = 30$ %, (d) $f_w = 40$ %.



Fig. S10 CD spectrum of *L*-GCS (black line) and *D*-GCS (red line) gel in MeCN/H₂O, (a) $f_w = 10$ %, (b) $f_w = 20$ %, (c) $f_w = 30$ %, (c) $f_w = 40$ %, [GCS] = 6.11 mM.



Fig. S11 CPL spectra of *L*-**GCS** (black line) and *D*-**GCS** (red line) in DMF, [**GCS**]=6.11 mM.



Fig. S12 SEM images of xerogels of GCS in (a) Toluene, (b) DMF/H₂O, $f_w = 40$ %, (c) CCl₄, [GCS]=6.11 mM.



Fig. S13 Solvent exchange experiments. (a) The organogel was firstly prepared in toluene and showed a bisignate Cotton effect (red dotted line). After removing toluene under vacuum, a xerogel was obtained, which was next dispersed in DMF/H₂O ($f_w = 40 \%$) mixture and formed organogel again (solid black line). (b) The procedures are the same as (a) but with opposite solvent sequence. Red dashed line: in DMF/H₂O gel, solid black line: in toluene gel.



Fig. S14 XRD pattern of the xerogel of toluene (red line) and DMF/H₂O co-solvents, $f_w = 40$ % (black line).



Fig. S15 FT-IR spectra of GCS in different solvents.



Fig. S16 Calculated CD spectrum of *L*-**GCS** with water-involved H-bonds by TD-DFT method.



Fig. S17 The fluorescence lifetime of GCS in different solvents, (a) DMF, (b) DMF/H₂O, $f_w = 40$ %, (c) toluene, (d) CCl₄, [GCS] = 6.11 mM, $\lambda_{ex} = 358.4$ nm.



3. NMR, MS spectra and quantum yields data for GCS.

Fig. S18 ¹H NMR spectrum for GCS (400 MHz, d_6 -DMSO, δ , ppm).



Fig. S19 ¹³C NMR spectrum for GCS (101 MHz, d_6 -DMSO, δ , ppm).



Fig. S20 MALDI-TOF-MS spectra for GCS.

4. Experimental Method.

The reagents and solvents were purchased from TCI and other companies and used without further purification.

Organogels Formation. A typical procedure for the gel formation in organic solution was as follows: 3 mg gelator were dispersed into 0.60 mL organic solvents and the mixture was heated to form a transparent solution then cooled to room temperature. Gels were obtained after 20 minutes.

Circular Dichroism (CD) spectra were recorded in quartzcurettes (light path 0.1 mm) on a JASCO J-810 spectrophotometer. The samples were take from the sample tube and cast on quartz plates without evaprating.

CPL measurements were performed with a JASCO CPL-200 spectrometer (light path 0.1 mm). The samples were prepared by directly taking gels from the test tube. In order to keep the detector at the best state and obtain the precise signals, the DV values were monitored to about 0.5 Voltage.

FT-IR spectra were recorded on a Bruker Tensor 27 FTIR spectrometer at room temperature. The KBr pellets made from the vacuum-dried samples of xeogels were used for FT-IR spectra measurements.

XRD analysis was performed on a Rigaku D/Max-2500 X-ray diffractometer (Japan) with Cu K α radiation ($\lambda = 1.5406$ Å), which was operated at a voltage of 40 kV and a

current of 200 mA. Samples were cast on glass substrates and vacuum-dried for XRD measurements.

Mass Spectra and Elemental Analysis. Mass spectral data were obtained by using a BIFLEIII matrix-assisted laser desorption/ionization time of fight mass spectrometry (MALDI-TOF MS) instrument and on an AB SCIEX 3200Q TRAP LC/MS/MS system equipped with an Agilent 1260 Infinity HPLC. Elemental analysis was performed on a Thermo Flash EA-1112 Series NCHS-O analyzer.

SEM images were recorded on a Hitachi S-4800 FE-SEM instrument with an accelerating voltage of 11 kV. Samples were cast onto single-crystal silica plates, the solvent was evaporated under the ambient conditions, and then vacuum-dried. The sample surface was coated with a thin layer of Pt to increase the contrast.

Quantum yield was measured by using an absolute PL quantum yield spectrometer (Edinburg FLS-980 fluorescence spectrometer) with a calibrated integrating sphere with Xe lamp $\lambda_{ex} = 340$ nm.

Fluorescence lifetime measurements were also recorded on the Edinburg FLS-980 fluorescence spectrometer using time-correlated single photon counting (TCSPC) with laser $\lambda_{ex} = 358.4$ nm.

5. Reference

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