Supporting Information

A novel aggregation-induced emission enhancement triggered by the assembly of chiral gelator: From non-emissive nanofibers to emissive micro-loops

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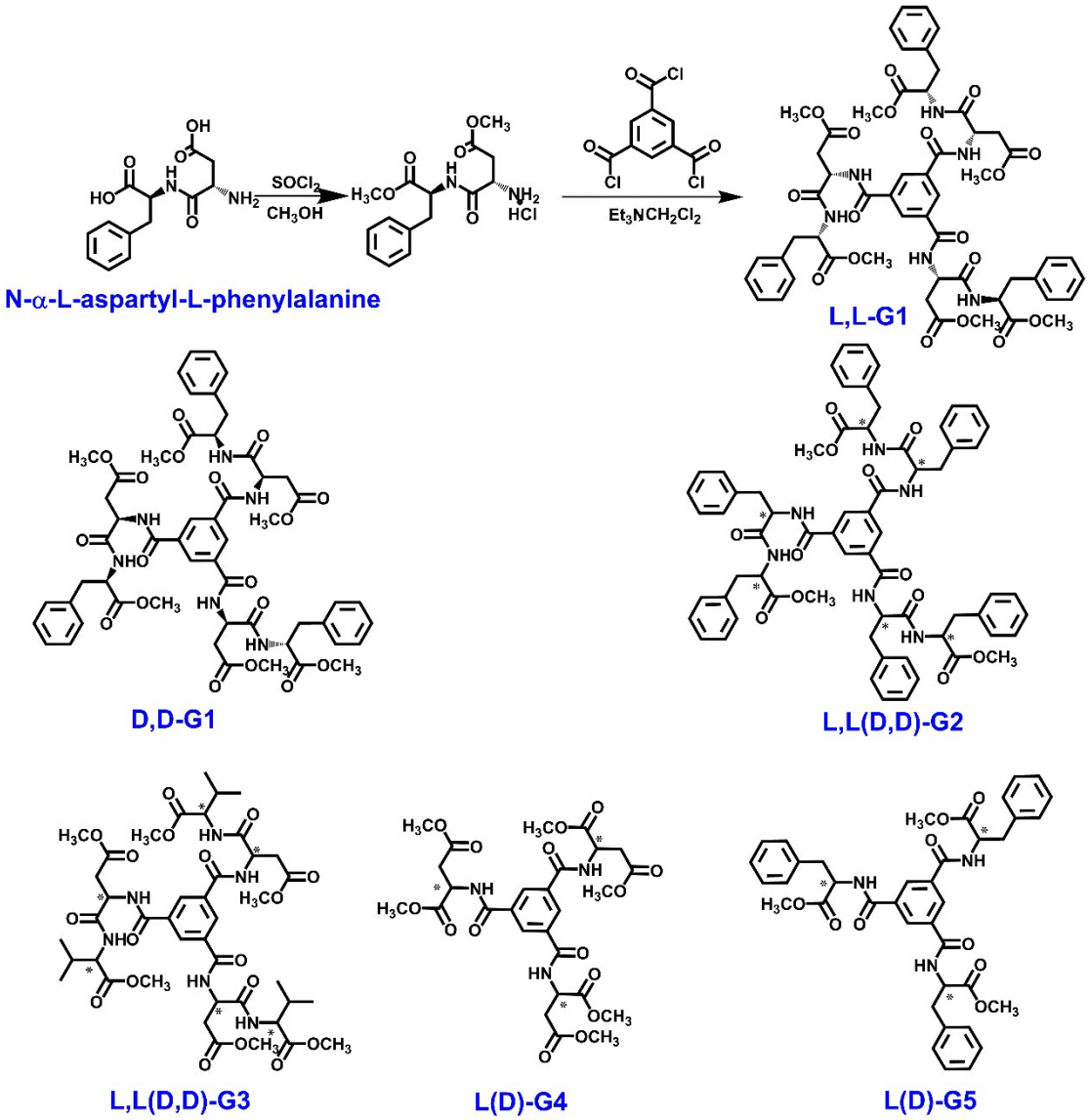
S1. Material and instruments.

1,3,5-benzenetricarboxylic acid chloride and thionyl chloride was purchased from Acros Organics (USA). L(D)-aspartylic acid and L(D)-phenylalanine were purchased from Aladdin Corp. (P.R. China). Triethylamine and deuterated dimethylsulfoxide (d_6-DMSO) were purchased from Sigma-Aldrich Corp. (P. R. China). Methanol, ethyl acetate with high purities (99.9%) were purchased from Sinopharm Chemical Reagent Corp. (P. R. China), and ethyl acetate was dried by molecular sieves for 24 hours prior to use. The L(D) dipeptides building blocks were synthesised using Fmoc-based peptide synthesis on the Wang resin through solid-supported peptide synthesis (CS-Bio Peptide Synthesizer CS 136XT (CA, USA).

Atomic Force Microscopy (AFM) investigation was conducted on a flat mica substrate using MultiMode VIII and Dimension FastScan Bio Scanning Probe Microscopy (Bruker, USA) with PeakForce QNM and tapping mode in air mode at 25 °C. Scanning Electron Microscope**(**SEM**)** images were recorded on Hitachi S-4800 Field Emission Scanning Electron Microscope at 10 KV accelerating voltage at 25°C. The SEM samples coated with a thin layer of gold of 3-5 nm using an E-1010 Ion Sputter (Hitachi). Fluorescence Microscope images were record on Zeiss Axio Observer A1 with filter set 02 (shift free (F) EX G 365, BS FT 395, EM LP 420). Fluorescence Spectra were recorded on a PerkinElmer LS55 fluorescence spectrophotometer. Dynamic Light Scattering (DLS) measurements were recorded using a Nano-ZS 90 Zetasizer of Malvern (UK) instrument. Both samples were measured at 25 °C directly. Infrared Spectra were performed with a Bruker Vertex 80v Fourier Transform Infrared Spectrometer (FT-IR) using ATR-DTGS at 25 °C. Nano FT-IR was performed with a scattering-type scanning near-field optical microscope s-SNOM (NeaSNOM, neaspec, Germany). ^{1}H NMR and ^{13}C NMR spectra were recorded on Bruker Advance 500 MHz spectrometer. Mass Spectra were obtained on a Thermo Scientific Q Exactive Plus quadrupole-orbitrap mass spectrometer. Circular Dichroism (CD) were recorded on JASCO J1500. Quartz cells with 1 cm lengths were used in CD measurements.

S2. Synthesis and characterization of L,L(D,D)-G1 to L(D)-G5



Scheme S1 Synthetic method of L,L-G1 (upper part). Chemical structures of D,D-G1 and other four pairs of gelators with different amino acid arms (lower part).

In general, the chiral dipeptides monomers were synthesized using standard 9-fluorenylmethoxycarbonyl (Fmoc) solid phase peptide synthesis on the 2 mmol scale using Fmoc-Phe(OtBu)-Wang resin. Fmoc deprotections were performed with 30% piperidine-DMF solution for 10 min. Amino acid coupling reactions were carried out using a coupling mixture of amino acid/HBTU/DIEA (4:3.95:6 relative to the resin) in the mixture of DCM/DMF. Cleavage of the peptides from the Wang resin was carried

out with a mixture of trifluoroacetic acid (TFA)/triisopropylsilane (TIS)/H_2O in a ratio of 95:2.5:2.5 for 3 hours. Excess TFA and scavengers were removed by rotary evaporation. The remaining peptide solution was triturated with cold ethyl ether and the precipitate was centrifuged, and the supernatant liquid was removed by decantation.

After washing with ethyl ether (three times) to remove residual TFA, the precipitate was dried under vacuum overnight. The peptides were purified by HPLC. Water/acetonitrile gradient containing 0.1 vol % TFA was used as an eluent at a flow rate of 4 mL/min. The purified fractions were collected and concentrated by rotary evaporation to remove acetonitrile, then lyophilized and stored at -20 °C.

The similar procedures can be used to synthesis the other five dipeptides.

2.1 Synthesis and characterization of L,L-G1:

To a solution in ice brine bath of N-α-L-aspartyl-L-phenylalanine (1.68 g, 6 mmol) in CH_3OH (30 mL), thionyl chloride (6 mL) was added dropwise for 30 minutes. After addition, the reaction mixture was stirred for 3 hours at room temperature. Most of solvent was evaporated under reduced pressure to gain the semi-oil residue. Then 30 mL fresh methanol was added to dissolve this residue and evaporated again under reduced pressure; repeated this process for five times to remove the extra thionyl chloride. Finally the residue was dried under vacuum to obtain pure white product (N-α-L-aspartyl-L-phenylalanine di-methyl ester hydrochloride).

Triethylamine (Et_3N) (1.01 g, 10 mmol) was added to a solution of N-α-L-aspartyl-L-phenylalanine di-methyl ester hydrochloride (2.07 g, 6 mmol) in 50 mL dry CH_2Cl_2, the mixture was stirred for 10 min, then 1,3,5-benzenetricarboxylic acid chloride (0.53 g, 2 mmol) in dry dichloromethane (DCM) (20 ml) was added dropwise to this mixture at ambient temperature, and continued to stir for 24 hours. The mixture was washed with water for three times, and the organic layer was dried over anhydrous sodium sulphate overnight. After filtration and evaporation of solvent, the crude product was purified on a silica gel column, with elution of CH_2Cl_2/CH_3OH (50:1), to give target compound as white solid L,L-G1(yield: 79%, m.p. 211 ºC). ^1H NMR (500 MHz, d_6-DMSO): δ 8.92 (d, J=7.8 Hz, 37H), 8.46 (s, 37H), 8.39 (d, J=7.5 Hz, 39H), 7.61-7.15 (m, 181H), 7.08 (dd, J=54.5, 6.4 Hz, 27H), 4.93 (dd, J=13.5, 8.3 Hz, 39H), 4.50 (dd,

J=13.7, 7.8 Hz, 39H), 3.66 (dd, J=18.8, 11.1 Hz, 11H), 3.60 (s, 224H), 3.60 (s, 232H), 3.33 (s, 406H), 3.06 (dd, J=19.2, 13.6 Hz, 21H), 3.00 (ddd, J=22.3, 13.8, 7.1 Hz, 81H), 2.92-2.69 (m, 81H), 2.53 (d, J=19.5 Hz, 98H), 1.24 (s, 6H), 0.10 (d, J=26.5 Hz, 12H), 0.01 (s, 472H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.1, 171.0, 165.9, 137.4, 134.7, 129.9, 129.5, 128.7, 127.0, 54.2, 52.4, 52.0, 50.4, 40.6-40.3, 40.1, 39.9, 39.7, 39.5, 39.5-39.3, 36.9, 35.9. IR: (3283, 2953, 1736, 1647, 1516, 1436, 1375, 1210, 1169, 993, 899, 743, 698, 485 cm^{-1}), ESI-MS: m/z calcd for $C_{54}H_{60}N_6O_{18}$: 1080.4; found: 1081.3 $[M+H]^+$. Elemental analysis calcd. (%) for $C_{54}H_{60}N_6O_{18}$: C, 59.99; H, 5.59; N, 7.77; O, 26.64; found: C, 59.72; H, 5.66; N, 7.74.

2.2 Synthesis and characterization of D,D-G1: N-α-D-aspartyl-D-phenylalanine di-methyl ester hydrochloride was prepared as a white solid according to the procedure similar to that for N-α-L-aspartyl-L-phenylalanine di-methyl ester hydrochloride. Triethylamine (Et_3N) (1.01 g, 10 mmol) was added to a solution of N-α-D-aspartyl-D-phenylalanine di-methyl ester hydrochloride (2.07 g, 6 mmol) in 50 mL dry CH_2Cl_2, the mixture was stirred for 10 min, then 1,3,5-benzenetricarboxylic acid chloride (0.53 g, 2 mmol) in dry dichloromethane (DCM) (20 ml) was added dropwise to this mixture at ambient temperature, and continued to stir for 24 hours. The mixture was washed with water for three times, and the organic layer was dried over anhydrous sodium sulphate overnight. After filtration and evaporation of solvent, the crude product was purified on a silica gel column, with elution of CH_2Cl_2/CH_3OH (50:1), to give target compound as white solid D,D-G1(yield: 78%, m.p. 210 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 8.92 (d, J=7.8 Hz, 37H), 8.46 (s, 37H), 8.39 (d, J=7.5 Hz, 39H), 7.61-7.15 (m, 181H), 7.08 (dd, J=54.5, 6.4 Hz, 27H), 4.93 (dd, J=13.5, 8.3 Hz, 39H), 4.50 (dd, J=13.7, 7.8 Hz, 39H), 3.66 (dd, J=18.8, 11.1 Hz, 11H), 3.60 (s, 224H), 3.60 (s, 232H), 3.33 (s, 406H), 3.06 (dd, J=19.2, 13.6 Hz, 21H), 3.00 (ddd, J=22.3, 13.8, 7.1 Hz, 81H), 2.92-2.69 (m, 81H), 2.53 (d, J= 19.5 Hz, 98H), 1.24 (s, 6H), 0.10 (d, J=26.5 Hz, 12H), 0.01 (s, 472H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.1, 171.0, 165.9, 137.4, 134.7, 129.9, 129.5, 128.7, 127.0, 54.2, 52.4, 52.0, 50.4, 40.6-40.3, 40.1, 39.9, 39.7, 39.5, 39.5-39.3, 36.9, 35.9. IR: (3283, 2953, 1736, 1647, 1516, 1436, 1375, 1210, 1169, 993, 899, 743, 698, 485 cm^{-1}), ESI-MS: m/z calcd for $C_{54}H_{60}N_6O_{18}$: 1080.4; found: 1081.3

[M+H]$^+$. Elemental analysis calcd. (%) for $C_{54}H_{60}N_6O_{18}$: C, 59.99; H, 5.59; N, 7.77; O, 26.64; found: C, 59.72; H, 5.66; N, 7.74.

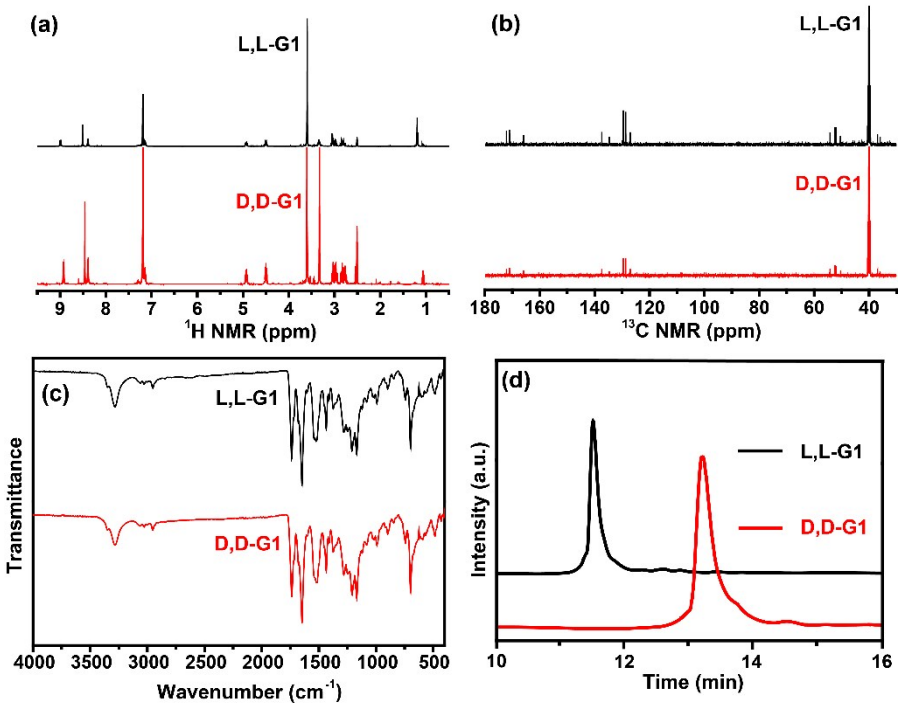


Fig. S1 Comparison of (a) ^{1}H NMR; (b) ^{13}C NMR; (c) FT-IR spectra; and (d) HPLC chiral chromatogram for the enantiomeric purities of L,L-G1 and D,D-G1. The spectra clearly indicated that the chemical structures of L,L-G1 and D,D-G1 were identical to each other except their molecular chirality.

2.3 Synthesis and characterization of L,L-G2: The same procedure was adopted to prepare L,L-G2 except the L-phenyl-L-phenylalanine was used as reactant (yield: 76%, m.p. 243 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 9.29-8.64 (m, 8H), 8.59 (d, J = 7.4 Hz, 8H), 8.23 (s, 7H), 7.55-5.79 (m, 84H), 7.15-7.09 (m, 3H), 7.06 (t, J=22.0 Hz, 5H), 7.06 (t, J=22.0 Hz, 2H), 6.98-6.08 (m, 1H), 4.83 (td, J=10.1, 4.3 Hz, 8H), 4.55 (dd, J=14.1, 7.7 Hz, 8H), 4.18 (d, J=234.0 Hz, 1H), 3.59 (s, 23H), 3.35 (s, 16H), 3.19-2.93 (m, 32H), 2.88 (d, J=8.1 Hz, 2H), 2.51 (s, 4H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.21, 171.84, 165.82, 138.51, 137.44, 134.77, 129.57, 128.74, 128.55, 127.06, 126.73, 54.92, 54.18, 52.34, 40.49, 39.99, 39.82, 39.59, 39.49, 37.62, 37.06. IR: (3149, 2964, 1788, 1610, 1587, 1485, 1329, 1211, 1186, 947, 832, 763, 628, 425 cm^{-1}). MS: m/z calcd for

$C_{66}H_{66}N_6O_{12}$: 1134.5; found: 1135.5. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{66}H_{66}N_6O_{12}$: C, 69.83; H, 5.86; N, 7.40; O, 16.91. Found: C, 69.88; H, 5.82; N, 7.49, O, 17.11.

2.4 Synthesis and characterization of D,D-G2: The same procedure was adopted to prepare D,D-G2 except the D-phenyl-D-phenylalanine was used as reactant (yield: 72%, m.p. 240 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 8.92-8.66 (m, 30H), 8.59 (d, J=7.4 Hz, 33H), 8.47-8.20 (m, 34H), 7.53-5.77 (m, 354H), 7.15 (dd, J=24.5, 19.0 Hz, 73H), 7.19 -5.77 (m, 81H), 7.09-5.93 (m, 8H), 4.83 (td, J=10.1, 4.3 Hz, 33H), 4.55 (dd, J=14.1, 7.7 Hz, 34H), 3.93 (d, J=16.3 Hz, 5H), 3.93 (d, J=16.3 Hz, 5H), 3.97-3.56 (m, 106H), 3.34 (s, 21H), 3.15-2.81 (m, 138H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.21, 171.84, 165.82, 138.51, 137.45, 134.77, 129.57, 128.64, 126.91, 126.73, 54.92, 54.18, 52.34, 40.43, 40.26, 40.10, 39.93, 39.76, 39.59, 39.50, 37.62. IR: (3149, 2964, 1788, 1610, 1587, 1485, 1329, 1211, 1186, 947, 832, 763, 628, 425 cm^{-1}). MS: m/z calcd for $C_{66}H_{66}N_6O_{12}$: 1134.5; found: 1135.5. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{66}H_{66}N_6O_{12}$: C, 69.83; H, 5.86; N, 7.40; O, 16.91. Found: C, 69.88; H, 5.82; N, 7.49, O, 17.11.

2.5 Synthesis and characterization of L,L-G3: The same procedure was adopted to prepare L,L-G3 except the N-α-L-aspartyl-L-valine was used as reactant (yield: 62%, m.p. 135 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 9.09 (d, J=7.0 Hz, 17H), 8.54 (s, 16H), 8.25 (d, J=8.1 Hz, 18H), 4.95 (q, J=7.2 Hz, 18H), 4.17 (dd, J=7.9, 6.3 Hz, 18H), 3.62 (d, J=10.3 Hz, 105H), 3.36 (d, J=19.6 Hz, 7H), 3.33 (s, 24H), 3.12-2.98 (m, 181H), 2.94-2.77 (m, 39H), 2.51 (d, J=1.6 Hz, 22H), 2.05 (dp, J=13.3, 6.6 Hz, 21H), 1.33 (t, J =7.3 Hz, 3H), 1.21 (t, J=7.3 Hz, 274H), 1.04 (ddd, J=25.7, 14.8, 7.3 Hz, 8H), 0.87 (t, J =7.6 Hz, 109H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.28, 171.15, 165.96, 134.70, 129.79, 58.01, 52.21, 52.01, 50.55, 45.77, 40.43, 40.34, 40.11, 40.00, 39.77, 39.67, 39.50, 35.93, 30.28, 19.41. IR: (3130, 2286, 1776, 1651, 1582, 1466, 1329, 1219, 1180, 911, 815, 716, 665, 435 cm^{-1}). MS: m/z calcd for $C_{42}H_{60}N_6O_{18}$: 936.4; found: 937.5. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{42}H_{60}N_6O_{18}$: C, 53.84; H, 6.45; N, 8.57; O, 30.79. Found: C, 53.87; H, 6.45; N, 8.48, O, 30.88.

2.6 Synthesis and characterization of D,D-G3: The same procedure was adopted to prepare D,D-G3 except the N-α-D-aspartyl-D-valine was used as reactant (yield: 60%, m.p. 133 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 9.06 (d, J=7.0 Hz, 17H), 8.58 (s, 16H), 4.95 (q, J=7.2 Hz, 18H), 4.17 (dd, J=7.9, 6.3 Hz, 18H), 3.66 (d, J=10.3 Hz, 105H), 3.35 (d, J=19.6 Hz, 7H), 3.33 (s, 24H), 2.94-2.77 (m, 39H), 2.51 (d, J=1.6 Hz, 22H), 2.05 (dp, J=13.3, 6.6 Hz, 21H), 1.36 (t, J =7.3 Hz, 3H), 1.04 (ddd, J=25.7, 14.8, 7.3 Hz, 8H), 0.87 (t, J =7.6 Hz, 109H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.8, 172.4, 171.8, 166.2, 137.1, 128.4, 57.9, 52.2, 51.9, 48.88, 34.9, 29.5, 19.0. IR: (3130, 2286, 1776, 1651, 1582, 1466, 1329, 1219, 1180, 911, 815, 716, 665, 435 cm^{-1}). MS: m/z calcd for $C_{42}H_{60}N_6O_{18}$: 936.4; found: 937.5. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{42}H_{60}N_6O_{18}$: C, 53.84; H, 6.45; N, 8.87; O, 30.84. Found: C, 53.87; H, 6.45; N, 8.48, O, 30.88.

2.7 Synthesis and characterization of L-G4: The same procedure was adopted to prepare L-G4 except the L-aspartylic acid was used as reactant (yield: 84%, m.p. 124 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 8.84-8.44 (m, 14H), 8.59-8.44 (m, 9H), 8.59-8.32 (m, 13H), 6.90 (s, 6H), 4.88-4.65 (m, 7H), 3.69 -3.39 (m, 55H), 3.39-3.29 (m, 4H), 3.00 (q, J=7.3 Hz, 177H), 2.95-2.70 (m, 32H), 2.70 -2.55 (m, 7H), 2.70-2.35 (m, 21H), 1.18 (t, J=7.3 Hz, 235H), 1.17-0.44 (m, 46H). ^{13}C NMR (126 MHz, d_6-DMSO): 173.1, 171.67, 166.2, 137.1, 128.4, 52.03, 51.86, 50.69, 45.64, 40.43, 40.27, 40.17, 40.00, 39.76, 39.67, 39.50. IR: (3184, 2248, 1763, 1672, 1582, 1443, 1313, 1225, 1188, 985, 715, 417 cm^{-1}). MS: m/z calcd for $C_{27}H_{33}N_3O_{15}$: 639.2; found: 640.2. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{27}H_{33}N_3O_{15}$: C, 50.68; H, 5.20; N, 6.57; O, 37.57. Found: C, 50.87; H, 5.45; N, 6.48, O, 37.65.

2.8 Synthesis and characterization of D-G4: The same procedure was adopted to prepare D-G4 except the D-aspartylic acid was used as reactant (yield: 84%, m.p. 125 ºC). ^{1}H NMR (500 MHz, d_6-DMSO): δ 8.86-8.48 (m, 34H), 8.37 (s, 12H), 6.90 (s, 6H), 5.03 (s, 3H), 4.87 (dd, J=13.9, 7.5 Hz, 16H), 3.69-3.33 (m, 390H), 3.28-2.80 (m, 334H), 2.73 (ddd, J=23.6, 14.3, 6.5 Hz, 27H), 2.51 (s, 17H), 1.20 (t, J=7.3 Hz, 388H), 1.09 (t, J=7.0 Hz, 282H). ^{13}C NMR (126 MHz, d_6-DMSO): 171.56, 171.38, 170.97, 170.07, 165.65, 134.41, 129.91, 52.69, 50.56, 49.89, 45.64, 40.49, 40.16, 39.99, 39.83, 39.66,

39.49, 35.84, 35.64. IR: (3184, 2248, 1763, 1672, 1582, 1443, 1313, 1225, 1188, 985, 715, 417 cm^{-1}). MS: m/z calcd for $C_{27}H_{33}N_3O_{15}$: 639.2; found: 640.2. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{27}H_{33}N_3O_{15}$: C, 50.71; H, 5.20; N, 6.59; O, 37.52. Found: C, 50.87; H, 5.45; N, 6.48, O, 37.65.

2.9 Synthesis and characterization of L-G5: The same procedure was adopted to prepare L-G5 except the L-phenylalanine was used as reactant (yield: 86%, m.p. 170 ºC). ^1H NMR (500 MHz, d_6-DMSO): δ 8.58-8.13 (m, 9H), 8.52-8.28 (m, 9H), 7.35-7.05 (m, 32H), 4.57 (s, 4H), 3.64 (s, 4H), 3.39 (q, J=7.0 Hz, 76H), 3.28-3.06 (m, 14H), 3.00 (q, J=7.1 Hz, 96H), 2.91-2.54 (m, 3H), 2.51 (d, J=1.5 Hz, 6H), 1.33 (ddd, J=24.3, 15.7, 8.4 Hz, 2H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.9, 166.1, 137.5, 137.1, 129.58, 128.72, 128.51, 126.58, 79.74, 65.38, 55.58-55.32, 45.66, 40.51, 40.27, 40.17, 40.00, 39.84, 39.60, 39.50, 37.11. IR: (3187, 2190, 1851, 1784, 1611, 1548, 1435, 1317, 1218, 1159, 822, 712 cm^{-1}). MS: m/z calcd for $C_{39}H_{39}N_3O_9$: 693.3; found: 694.4. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{39}H_{39}N_3O_9$: C, 67.50; H, 5.67; N, 6.05; O, 20.76. Found: C, 67.87; H, 5.45; N, 6.48; O, 20.78

2.10 Synthesis and characterization of D-G5: The same procedure was adopted to prepare D-G5 except the D-phenylalanine was used as reactant (yield: 88%, m.p. 170 ºC). ^1H NMR (500 MHz, d_6-DMSO): δ 8.55- 8.30 (m, 8H), 7.38-7.08 (m, 20H), 6.65 (s, 6H), 4.81 (s, 3H), 4.64 (ddd, J=21.5, 13.7, 9.0 Hz, 3H), 3.65 (s, 7H), 3.39 (q, J=7.0 Hz, 144H), 3.29-2.93 (m, 56H), 2.93-2.12 (m, 4H), 2.60-2.12 (m, 2H), 2.54-2.47 (m, 3H). ^{13}C NMR (126 MHz, d_6-DMSO): 172.41, 166.1, 137.5, 137.1, 129.56, 128.66 (s), 128.48, 126.90, 79.72, 65.38, 65.22, 55.10, 52.37, 45.70, 40.43, 40.28, 40.17, 39.94, 39.83, 39.67, 39.50, 36.57. IR: (3187, 2190, 1851, 1784, 1611, 1548, 1435, 1317, 1218, 1159, 822, 712 cm^{-1}). MS: m/z calcd for $C_{39}H_{39}N_3O_9$: 693.3; found: 694.4. $[M+H]^+$. Elemental analysis calcd. (%) for $C_{39}H_{39}N_3O_9$: C, 67.52; H, 5.67; N, 6.06; O, 20.76. Found: C, 67.87; H, 5.45; N, 6.48; O, 20.78

S3. Macroscopic organogel formation and self-assembled morphologies of L,L(D,D)-G1 aggregates in methanol.

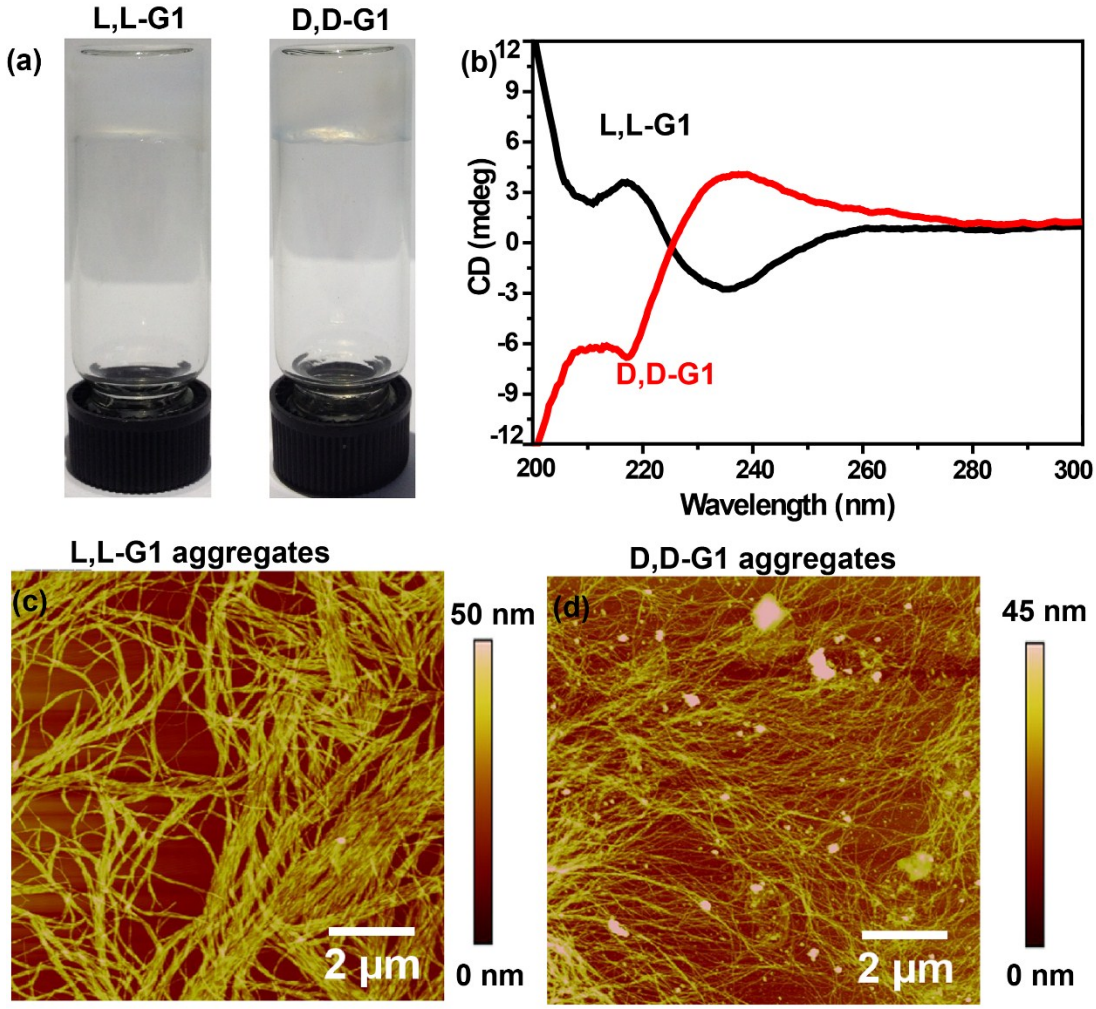


Fig. S2 Macroscopic organogel images (a), CD spectra (b, 10^{-5} M in methanol) and self-assembled morphologies in methanol: L,L-G1 aggregates (c) and D,D-G1 aggregates (d). According to Fig. a to d, both of L,L-G1 and D,D-G1 formed macroscopic organogel in methanol with with their microstructures containing three-dimensional cross-linked fibers.

S4. Self-assembled morphologies of L,L(D,D)-G1 on the mica surface at different concentrations in ethyl acetate.

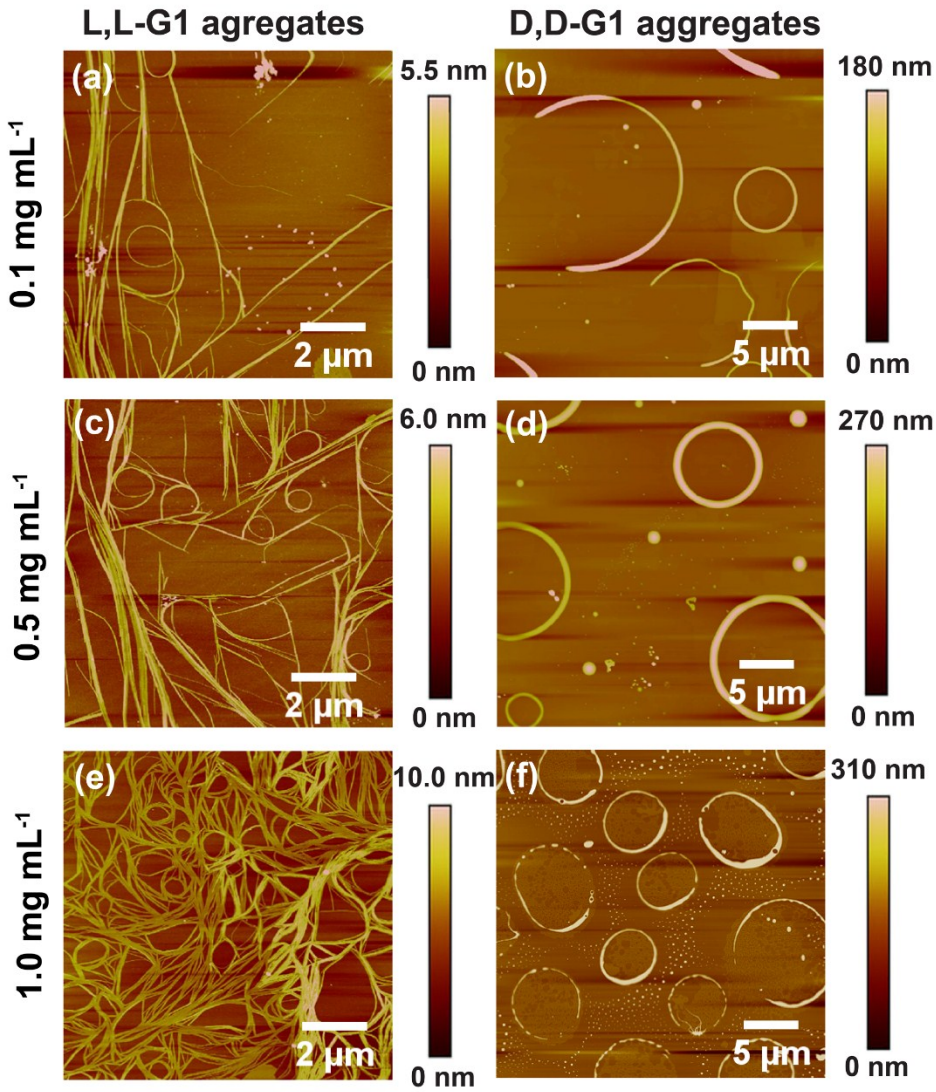


Fig. S3 Self-assembled morphologies of L,L-G1 (a, c, e) and D,D-G1 (b, d, f) in ethyl acetate on freshly cleaved mica surface: (a, b) 0.1, (c, d) 0.5, (e, f) 1.0 mg mL^{-1}, observed by AFM in the tapping mode. According to Fig. a-f, dynamic self-assembly growth of L,L-G1 and D,D-G1 would happen at low, medium, and high concentrations, 0.5 mg mL^{-1} was chosen for following research.

S5. Self-assembled morphologies of L,L(D,D)-G1 observed by SEM in a large scale.

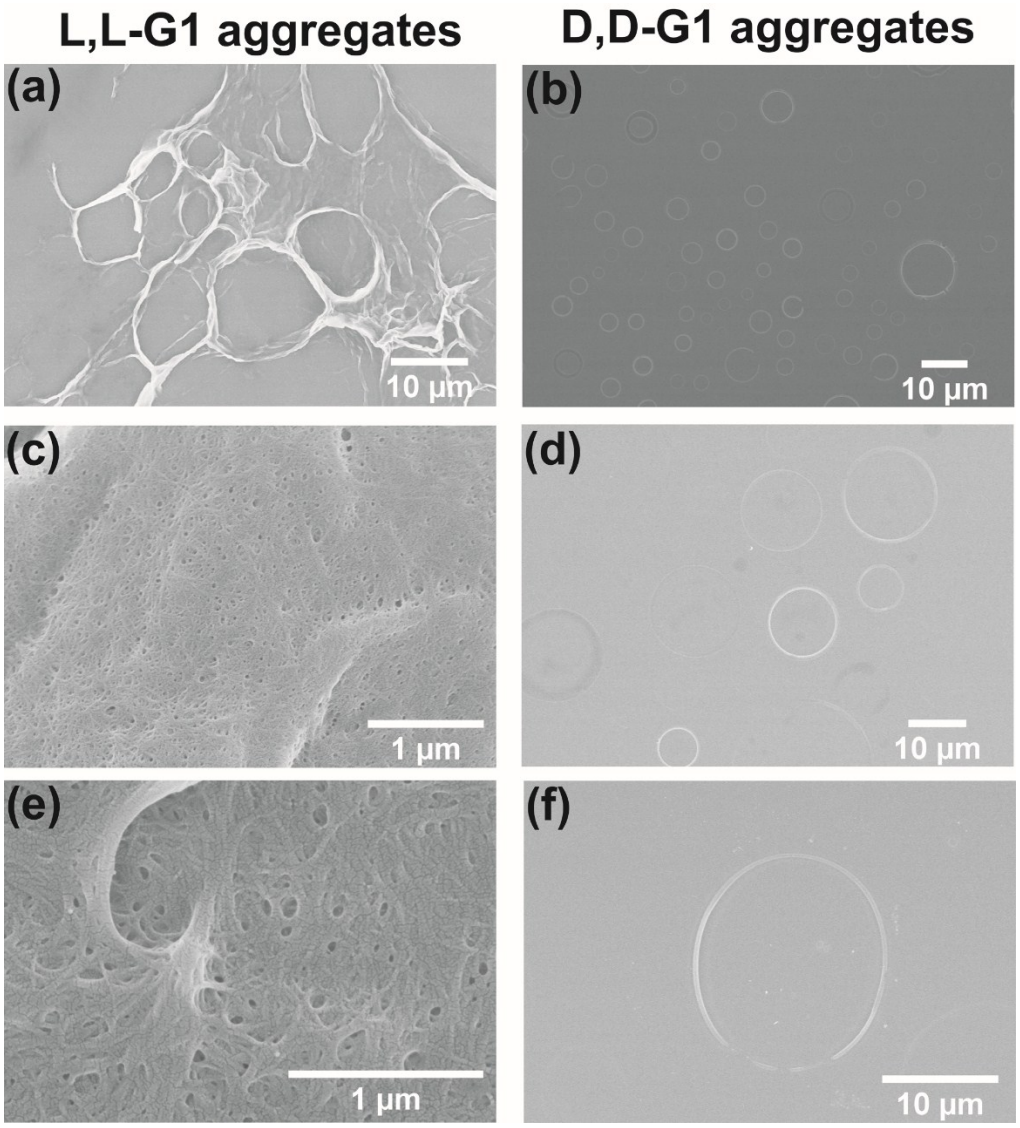


Fig. S4 Self-assembled morphologies of L,L-G1 (a, c, e) and D,D-G1 (b, d, f) observed by SEM. These data indicated that significant chiral self-assembled discrimination could also be observed in a large scale.

S6. Self-assembled morphologies of mixed L,L(D,D)-G1 aggregates with different ratio.

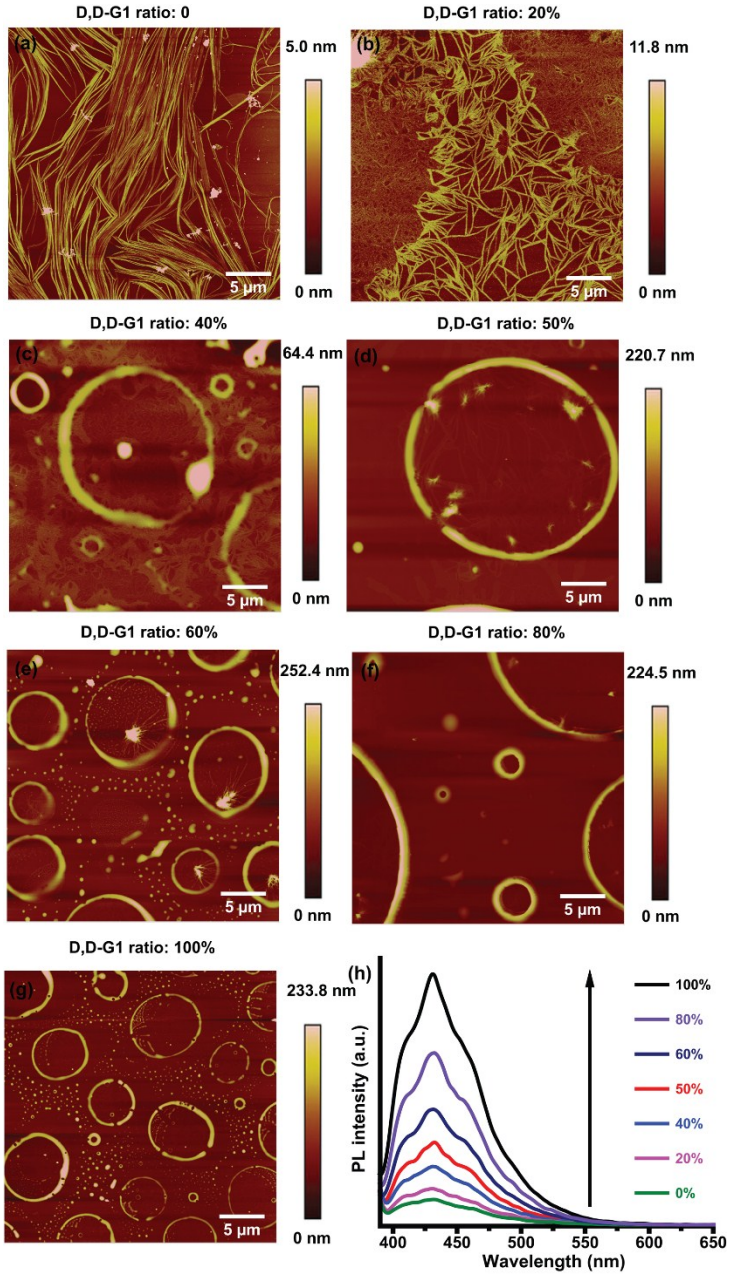


Fig. S5 AFM images of self-assembly nanostructure of mixture L,L-G1 and D,D-G1. D,D-G1 ratio:(a) 0; (b) 20%; (c) 40%; (d) 50%; (e) 60%; (f) 80%; (g) 100%. PL spectra of mixture of L,L-G1 and D,D-G1 with the increase ratio of D,D-G1. (Concentration: 10^{-4} M, λ_{ex}: 370 nm)

S7. Self-assembled morphologies of L,L(D,D)-G2 to L(D)-G5 aggregates observed by AFM.

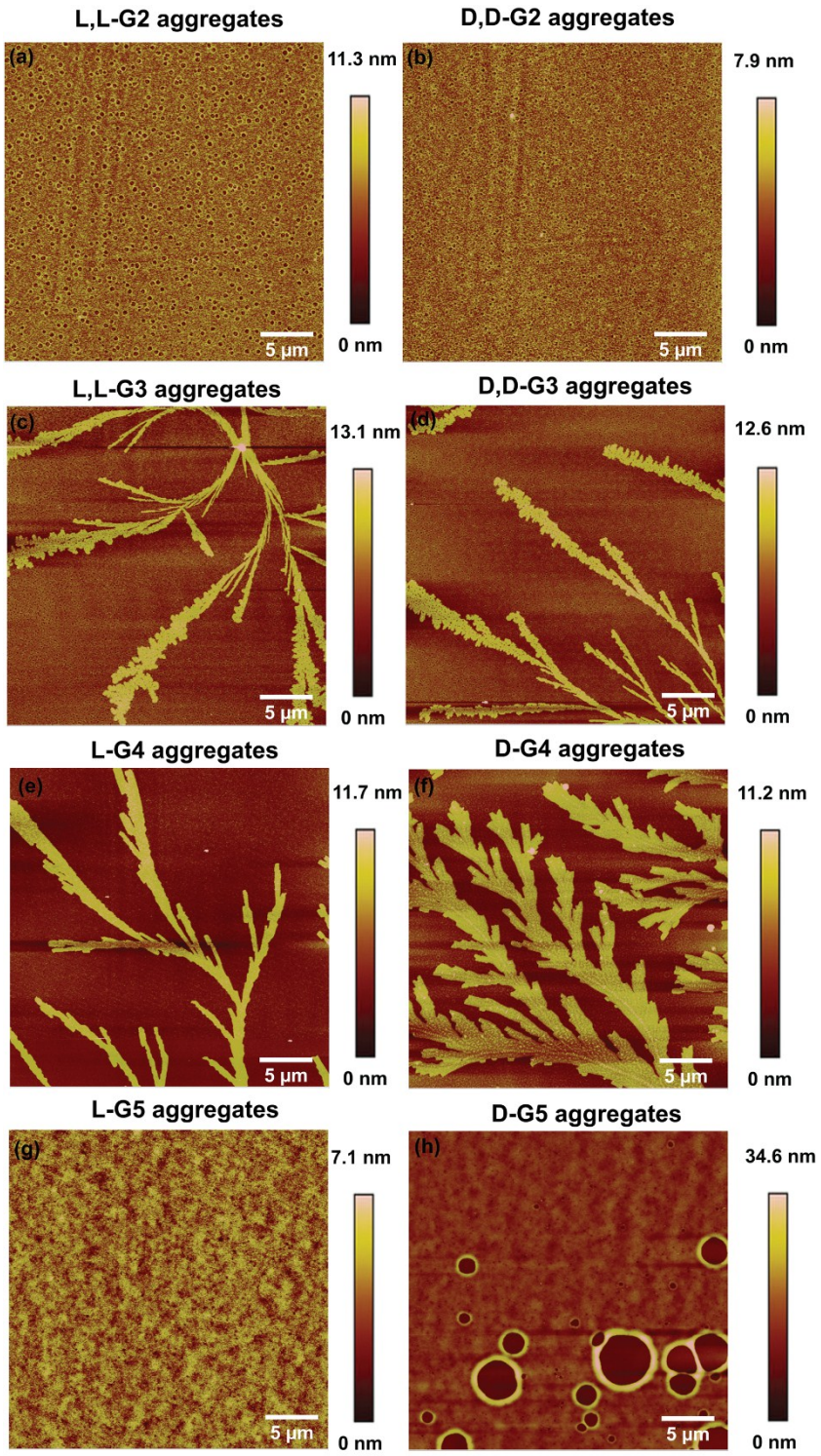


Fig. S6 AFM images of four pairs of compounds self-assembly nanostructure: L,L(D,D)-G2 (a,b); L,L(D,D)-G3(c,d); L(D)-G4(e,f) and L(D)-G5 (g,h).

S8. ATR-FTIR, Nano-FTIR and s-SNOM test of L,L-G1 and D,D-G1 aggregates.

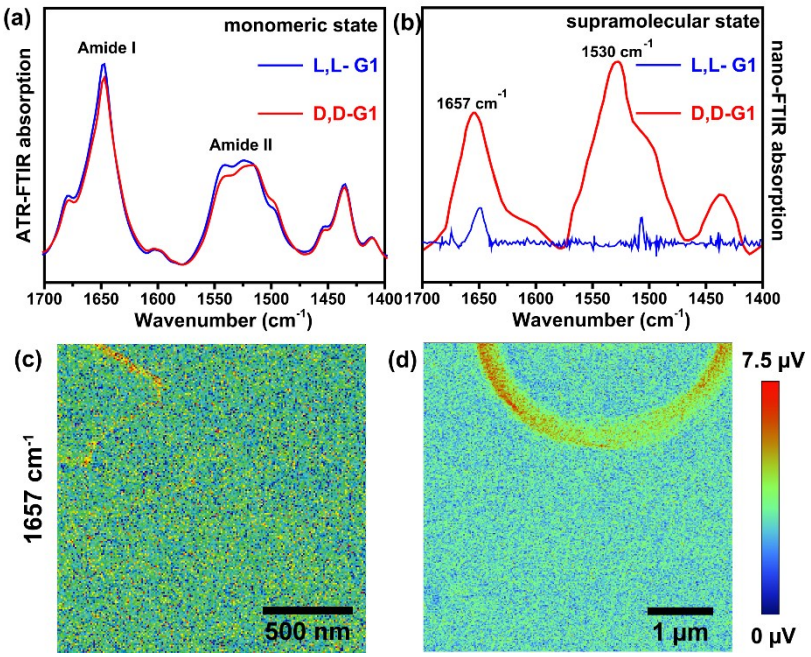


Fig. S7 ATR-FTIR spectrum of monomeric samples (a); Nano-FTIR spectra of self-assembled superstructures (2 min total acquisition time, 10 cm^{-1} resolution) (b); and infrared near-field images of aggregates at 1657 cm^{-1}: (c) L,L-G1 (d) D,D-G1.