Supporting Information for

Heterogeneous H-Bonding in a Foldamer Helix

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I. Materials and Instrumentation

Materials and Reagents

Most solvents and reagents used were obtained from Sigma-Aldrich. Amino acids and coupling reagents were obtained from Chem-Impex. International, Inc. Silica column chromatography was performed using Sigma-Aldrich technical grade silica gel, pore size 60 Å, 40-63 μm mesh size. Automated silica column chromatography was performed on a Biotage SP1 HPFC system using 10g SNAP cartridges. TLC was performed using SiliCycle Siliaplate glass-backed TLC plates, 60 Å pore size, 250 μm thickness, F-254 indicator. Visualization of TLC plates was performed with 254 nm UV light, ninhydrin stain (1.5 g ninhydrin in 100 mL butanol and 3 mL AcOH), $KMnO_4$ stain (1.5 g $KMnO_4$, 10 g K_2CO_3, and 1.25 mL 10% NaOH in 200 mL H_2O), PMA stain (10% PMA in EtOH), or I_2 stain (a few crystals of I_2 in a jar of silica gel).

NMR Spectrometers

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NMR Spectrometer	Grant	Award year
Bruker AC-300	NSF CHE-9208463	1993
	NIH S10 RR08389-01	1993
Varian Mercury-300	NSF CHE-034299	2003
Varian INOVA-600	NIH 1 S10 RR13866-01	2000
Bruker Avance III-400	NSF CHE-1048642	2010
Bruker Avance III-500	Generous gift from Paul J. Bender	2012

Mass Spectrometers

Mass Spectrometer	Grant	Award year
Bruker REFLEX-II MALDI	NSF CHE-9520868	1995
Waters Micromass LCT	NSF CHE-9974839	1999

II. Synthesis

a. General Procedures/Nomenclature

1. **General procedure 1:** Amide coupling

   Boc-protected amino acid or peptide is pre-activated by stirring at 0.1M in DMF or CH_2Cl_2 with coupling reagent (EDCI-HCl), coupling additive (HOBt, HOAt, Oxyma Pure, or DMAP), and DIEA for 15 min. Mixture is added to Boc-deprotected α- or γ-amino ester and stirred overnight. After completion, mixture is diluted with EtOAc and the organic phase is washed with 0.5 M HCl, 5% LiCl, saturated aqueous $NaHCO_3$, and brine. The

organic phase is dried with Na_2SO_4, concentrated *in vacuo*, and crude product is purified by silica column chromatography.

2. **General procedure 2:** Boc deprotection

   To Boc-protected amino ester or peptide is added 4.0 M HCl in dioxane. Mixture is stirred until completion as determined by TLC, and solvent is evaporated under a stream of N_2. Crude product is diluted with CH_2Cl_2 and solvent is removed again (2x). Remaining CH_2Cl_2 is removed under high vacuum and crude product is used without further purification.

3. **General procedure 3:** Benzyl ester hydrogenolysis

   Boc-, benzyl ester-protected γ-amino acid or peptide is dissolved in EtOH to 0.1 M. 10 wt% Pd/C is added (≈10 mg Pd/C per 30 mg amino acid) under N_2. The reaction vessel is purged of air in vacuo and refilled with N_2. The reaction vessel is equipped with a balloon of H_2 and stirred at room temperature for 5 hr. The reaction mixture is filtered through a bed of Celite and concentrated *in vacuo* to produce a white solid, which is used without further purification.

b. Compound Nomenclature Notes

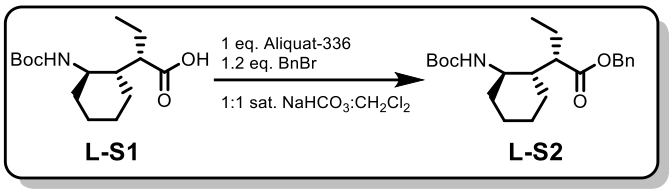
Abbrev.	**Full name**
EDCI-HCl	N-(3-dimethylaminopropyl)-N-ethylcarbodiimide hydrochloride
HOBt	1-hydroxybenzotriazole
Oxyma Pure	ethyl 2-cyano-2-(hydroxyimino)acetate
DIEA	diisopropylethylamine
DMF	dimethylformamide
EtOAc	ethyl acetate
HOAt	1-hydroxy-7-azabenzotriazole
HATU	1-[bis(dimethylamino)methylene]-1H-1,2,3-triazolo[4,5-b]pyridinium 3-oxid hexafluorophosphate
DMAP	4-dimethylaminopyridine

Compounds not appearing in the main text are numbered separately, with the letter "S" preceding the numbering of these compounds.

Only syntheses of one enantiomer are described for compounds in which both enantiomers were used. For simplicity, the enantiomer is designated by the configuration of the α-amino acid in the α/γ(**I**)-peptide that incorporates that compound. Thus, "**L-xx**" would be compound number **xx** that would be used in an α/γ(**I**)-peptide incorporating an L-α-amino acid and (*S*,*S*,*R*)-γ(**I**), and "**D-yy**" would be compound number **yy** that would be used in an α/γ(**I**)-peptide incorporating a D-α-amino acid and (*R*,*R*,*S*)-γ(**I**).

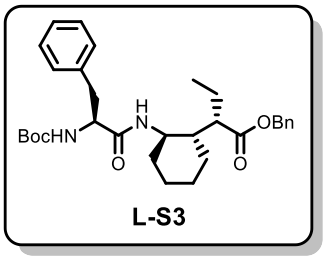
c. Synthesis

L-S2 – Boc-[(*S*,*S*,*R*)-EtACHA]-OBn



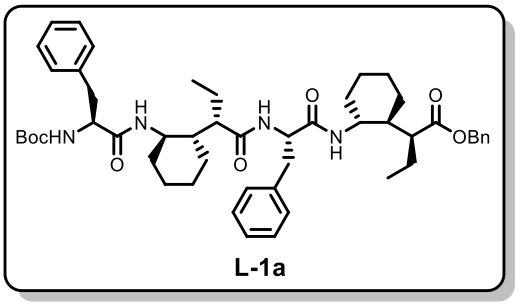
Compound **L-S2** was synthesized according to previously reported methods.[S1] Boc-amino acid **L-S1** (1.14 g, 3.98 mmol) was dissolved in a 1:1 binary mixture of CH_2Cl_2 and saturated aqueous $NaHCO_3$. Aliquat-336 (1 eq., 1.61 g, 3.98 mmol) and benzyl bromide (1.2 eq., 568 µL, 4.78 mmol) were added, and the reaction mixture was stirred at room temperature for 16 hr. The reaction mixture was extracted three times with CH_2Cl_2, and the combined organic layers were washed with brine. The organic layer was dried over Na_2SO_4, filtered, and concentrated *in vacuo* to yield a pale yellow viscous oil. This residue was purified *via* silica column chromatography eluting with 7:1 hexanes/EtOAc to yield 1.33 g **L-S2** as a colorless viscous oil (3.54 mmol, 89% yield). **TLC** R_f = 0.47 (4:1 hexanes/EtOAc, ninhydrin stain) **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.41 – 7.28 (m, 5H), 5.16 (d, *J* = 12.4 Hz, 1H), 5.11 (d, *J* = 12.6 Hz, 1H), 4.33 (d, *J* = 10.0 Hz, 1H), 3.39 (qd, *J* = 10.6, 3.7 Hz, 1H), 2.50 (dt, *J* = 9.1, 4.6 Hz, 1H), 2.03 – 1.92 (m, 1H), 1.77 – 1.51 (m, 5H), 1.42 (s, 9H), 1.35 – 0.95 (m, 5H), 0.85 (t, *J* = 7.3 Hz, 3H). Small peaks (integration ≈ 0.15) were identified as rotamers with gradient 1D-NOESY with 2 s mixing time.[S2] **HRMS** *m/z* (ESI): calc. for $[C_{22}H_{33}NNaO_4]^+$ ($[M+Na]^+$) 398.2302, found 398.2318.

L-S3 – Boc-[^{L}Phe][(*S*,*S*,*R*)-EtACHA]-OBn



Boc-, benzyl ester-protected cyclic γ-amino ester **L-S2** (1.33 g, 3.54 mmol) was deprotected at the N-terminus according to **General Procedure 2**. The amine-deprotected salt was coupled according to **General Procedure 1** to Boc-L-phenylalanine (2 eq., 1.88 g, 7.08 mmol) using EDCI-HCl (2 eq., 1.36 g, 7.08 mmol), HOBt (2 eq., 1.28 g, 7.08 mmol), and DIEA (6 eq., 3.70 mL, 21.2 mmol) stirring at room temperature for 16 hr. Crude yellow viscous oil product was purified by silica column chromatography eluting with 4:1 hexanes/EtOAc to yield **L-S3** as 1.61 g white foam (3.09 mmol, 87% yield). **TLC** R_f = 0.36 (3:1 hexanes/EtOAc). **^{1}H NMR** (300 MHz, $CDCl_3$) δ 7.42 – 7.12 (m, 10H), 5.43 (d, *J* = 9.6 Hz, 1H), 5.12 (d, *J* = 12.3 Hz, 1H), 5.07 (d, *J* = 12.5 Hz, 1H), 4.20 (q, *J* = 8.2 Hz, 1H), 3.68 (q, *J* = 9.9 Hz, 1H), 3.12 – 2.89 (m, 2H), 2.37 (dd, *J* = 10.5, 3.7 Hz, 1H), 1.68 – 1.47 (m, 7H), 1.38 (s, 9H), 1.31 – 0.91 (m, 5H), 0.81 (t, *J* = 7.3 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{31}H_{43}N_2O_5]^+$ ($[M+H]^+$) 523.3167, found 523.3154.

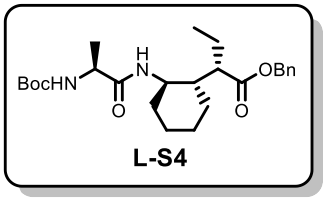
L-1a – Boc-([^{L}Phe][(*S*,*S*,*R*)-EtACHA])$_2$-OBn



L-1a

Boc-, benzyl ester-protected dipeptide **L-S3** (523 mg, 1.00 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, another amount of **L-S3** (1 eq., 523 mg, 1.00 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1 eq., 192 mg, 1.00 mmol), HOAt (1 eq., 1.68 mL from 0.6 M in DMF stock solution, 1.00 mmol), and DIEA (3 eq., 523 μL, 3.00 mmol) stirring at room temperature for 16 hr. Purification was carried out with silica column chromatography eluting with 2:1 hexanes/EtOAc to yield **L-1a** as 427 mg white foam (0.510 mmol, 51% yield). **TLC** R_f = 0.22 (2:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.40 (d, *J* = 7.7 Hz, 1H), 7.38 – 7.04 (m, 15H), 6.28 – 6.23 (m, 1H), 5.56 (d, *J* = 9.6 Hz, 1H), 5.12 (d, *J* = 12.3 Hz, 2H), 5.07 (d, *J* = 12.5 Hz, 1H), 4.49 (q, *J* = 7.8 Hz, 1H), 4.17 (q, *J* = 9.7, 8.6 Hz, 1H), 3.66 (qd, *J* = 10.6, 3.7 Hz, 1H), 3.57 (qd, *J* = 11.0, 4.0 Hz, 1H), 3.13 – 2.88 (m, 4H), 2.44 (d, *J* = 10.9 Hz, 1H), 2.18 (d, *J* = 11.6 Hz, 1H), 1.90 (d, *J* = 10.9 Hz, 1H), 1.66 – 1.45 (m, 11H), 1.39 (s, 9H), 1.31 – 0.85 (m, 10H), 0.81 (t, *J* = 7.2 Hz, 3H), 0.73 (t, *J* = 7.1 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{50}H_{69}N_4O_7]^+$ ($[M+H]^+$) 837.5161, found 837.5138.

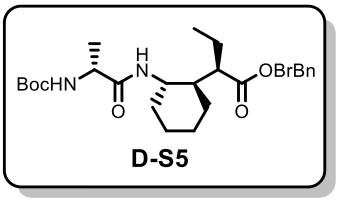
L-S4 – Boc-[^{L}Ala][(*S*,*S*,*R*)-EtACHA]-OBn



L-S4

L-Alanine benzyl ester 4-toluenesulfonate salt (85 mg, 0.24 mmol) was coupled according to **General Procedure 1** to Boc-L-alanine (4.1 eq., 2.36 g, 12.5 mmol) using EDCI-HCl (3.6 eq., 2.08 g, 10.9 mmol), HOAt (3.6 eq., 18.1 mL from 0.6 M in DMF stock solution, 10.9 mmol), and DIEA (3.6 eq., 1.89 mL, 10.9 mmol) stirring at room temperature for 12 hr. Crude yellow viscous oil product was purified by silica column chromatography eluting with 3:1→2:1 hexanes/EtOAc to yield **L-S4** as 1.01 g white foam (2.26 mmol, 75% yield). **TLC** R_f = 0.34 (2:1 hexanes/EtOAc). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.43 – 7.28 (m, 5H), 5.91 (d, *J* = 9.7 Hz, 1H), 5.13 (d, *J* = 12.4 Hz, 1H), 5.10 (d, *J* = 12.4 Hz, 1H), 5.03 – 4.93 (m, 1H), 4.10 – 4.01 (m, 1H), 3.76 (qd, *J* = 10.7, 4.0 Hz, 1H), 2.42 (d, *J* = 10.4 Hz, 1H), 1.97 – 1.88 (m, 1H), 1.75 – 1.46 (m, 7H), 1.41 (s, 9H), 1.35 – 1.05 (m, 6H), 0.81 (t, *J* = 7.3 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{25}H_{38}N_2O_5Na]^+$ ($[M+Na]^+$) 469.2673, found 469.2664.

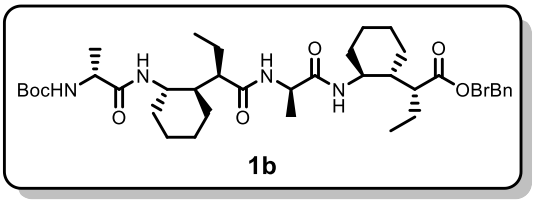
D-S5 – Boc-[^{D}Ala][(*R*,*R*,*S*)-EtACHA]-OBrBn



D-S5

Boc-, benzyl ester-protected dipeptide **D-S4** (186 mg, 0.416 mmol) was benzyl deprotected at the C-terminus according to **General Procedure 3**. To benzyl ester-deprotected product dissolved to 0.1 M in DMF as added Cs_2CO_3 (1.2 eq., 163 mg, 0.499 mmol) and 4-bromobenzyl bromide (1.2 eq., 125 mg, 0.499 mmol). The mixture was stirred for 16 hours, then diluted with EtOAc, the organic phase washed with 10% aqueous citric acid, saturated aqueous $NaHCO_3$, and brine. The organic phase was concentrated *in vacuo* to yield 189 mg crude **D-S5** (0.36 mmol, 87% yield) which was used without further purification. **TLC** R_f = 0.60 (1:1 hexanes/EtOAc).

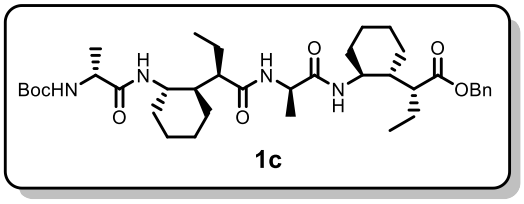
1b – Boc-([^{D}Ala][(*R*,*R*,*S*)-EtACHA])$_2$-OBrBn



1b

Boc-, benzyl ester-protected dipeptide **D-S5** (189 mg, 0.36 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, **D-S4** (1 eq., 161 mg, 0.36 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.2 eq., 84 mg, 0.44 mmol), HOBt (1.2 eq., 60 mg, 0.44 mmol), and DIEA (1.5 eq., 94 μL, 0.54 mmol) stirring at room temperature for 24 hr. Crude product was purified via silica column chromatography eluting with 1:1 EtOAc:hexanes to yield 220 mg **1b** as a white solid (80% yield). **TLC** R_f = 0.18 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (300 MHz, $CDCl_3$) δ7.49 (m, 2H), 7.29 (d, *J* = 7.4 Hz, 1H), 7.20 (m, 2H), 6.62 (d, *J* = 9.5 Hz, 1H), 6.03 (d, *J* = 9.6 Hz, 1H), 5.13, 5.10 (AB, J_{AB} = 12.7 Hz, 2H), 4.99 (d, *J* = 6.7 Hz, 1H), 4.46 (p, *J* = 7.2 Hz, 1H), 3.84 (p, *J* = 7.0 Hz, 1H), 3.71 (qd, *J* = 10.3, 3.8 Hz, 2H), 2.48 (m, 1H), 2.27 (m, 1H), 2.08 (m, 1H), 2.04-1.01 (m, 26H), 0.82 (m, 6H). **HRMS** *m/z* (ESI): calc. for $[C_{38}H_{59}BrN_4O_7Na]^+$ ($[M+Na]^+$) 786.8355, found 786.8326.

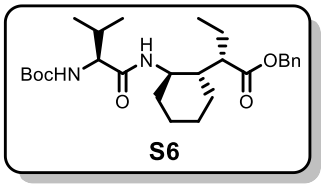
1c – Boc-([^{D}Ala][(*R*,*R*,*S*)-EtACHA])$_2$-OBn



1c

Boc-, benzyl ester-protected dipeptide **D-S5** (357 mg, 0.80 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, another amount of **D-S5** (1 eq., 357 mg, 0.80 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.2 eq., 184 mg, 0.96 mmol), HOBt (1.2 eq., 130 mg, 0.96 mmol), and DIEA (2 eq., 280 μL, 1.61 mmol) stirring at room temperature for 24 hr. Crude product was purified via silica column chromatography eluting with 1:1 EtOAc:hexanes to yield 438 mg **1b** as a white solid (80% yield). **TLC** R_f = 0.19 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.42 – 7.29 (m, 5H), 6.74 (d, *J* = 9.7 Hz, 1H), 6.09 (d, *J* = 9.5 Hz, 1H), 5.20 (d, *J* = 12.6 Hz, 1H), 5.16 (d, *J* = 12.7 Hz, 1H), 5.03 (d, *J* = 6.5 Hz, 1H), 4.49 (p, *J* = 7.3 Hz, 1H), 3.84 (p, *J* = 7.0 Hz, 1H), 3.79 – 3.58 (m, 2H), 2.52 – 2.45 (m, 1H), 2.29 (d, *J* = 11.3 Hz, 1H), 2.10 (d, *J* = 13.0 Hz, 1H), 2.00 (d, *J* = 12.7 Hz, 1H), 1.92 (d, *J* = 10.2 Hz, 1H), 1.83 – 1.50 (m, 12H), 1.40 (s, 9H), 1.38 – 1.03 (m, 14H), 0.84 (t, *J* = 7.3 Hz, 3H), 0.78 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{38}H_{60}N_4O_7Na]^+$ ($[M+Na]^+$) 707.4355, found 707.4326.

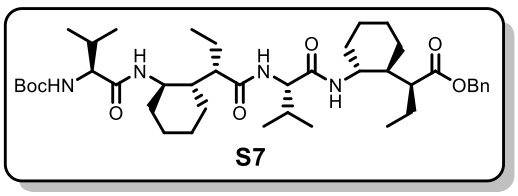
S6 – Boc-[Val][(*S,S,R*)-EtACHA]-OBn



S6

Boc-, benzyl ester-protected cyclic γ-amino ester **L-S2** (1.13 g, 3.00 mmol) was deprotected at the N-terminus according to **General Procedure 2**. The amine-deprotected salt was coupled according to **General Procedure 1** to Boc-L-valine (3.6 eq., 2.36 g, 10.9 mmol) using EDCI-HCl (3.6 eq., 2.08 g, 10.9 mmol), HOAt (3.6 eq., 18.1 mL from 0.6 M stock solution in DMF, 10.9 mmol), and DIEA (3.6 eq., 1.89 mL, 10.9 mmol) stirring at room temperature for 16 hr. Crude yellow viscous oil product was purified by silica column chromatography eluting with 4:1 hexanes/EtOAc to yield 1.42 g white solid **S6** (2.98 mmol, 99% yield). **TLC** R_f = 0.53 (2:1 hexanes/EtOAc). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.40 – 7.28 (m, 5H), 5.81 (d, *J* = 9.6 Hz, 1H), 5.18 – 5.03 (m, 3H), 3.86 – 3.69 (m, 2H), 2.45 (dt, *J* = 10.9, 3.4 Hz, 1H), 2.21 – 2.06 (m, 1H), 1.99 – 1.89 (m, 1H), 1.77 – 1.41 (m, 5H), 1.40 (s, 9H), 1.34 – 1.01 (m, 5H), 0.94 (d, *J* = 6.7 Hz, 3H), 0.91 (d, *J* = 6.9 Hz, 3H), 0.81 (t, *J* = 7.3 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{27}H_{42}N_2NaO_5]^+$ ($[M+Na]^+$) 497.2986, found 497.2986.

S7 – Boc-([^{L}Val][(*S,S,R*)-EtACHA])$_2$-OBn

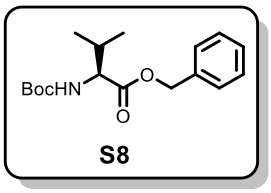


S7

Boc-, benzyl ester-protected dipeptide **S6** (528 mg, 1.11 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, another amount of **S6** (1.2 eq., 632 mg,

1.33 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.2 eq., 255 mg, 1.33 mmol), HOAt (1.2 eq., 2.22 mL from 0.6 M in DMF stock solution, 1.33 mmol), and DIEA (2 eq., 387 µL, 2.22 mmol) stirring at room temperature for 16 hr. Purification was carried out with silica column chromatography eluting with 2:1 hexanes/EtOAc to yield 547 mg **S7** as a white foam (0.738 mmol, 66% yield). **TLC** R_f = 0.49 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.43 – 7.27 (m, 5H), 7.11 (d, *J* = 8.6 Hz, 1H), 6.46 (d, *J* = 9.6 Hz, 1H), 5.84 (d, *J* = 9.5 Hz, 1H), 5.17 (d, *J* = 12.5 Hz, 1H), 5.13 (d, *J* = 12.6 Hz, 1H), 5.00 (d, *J* = 7.9 Hz, 1H), 4.20 – 4.11 (m, 1H), 3.83 – 3.66 (m, 2H), 3.63 (t, *J* = 7.7 Hz, 1H), 2.50 (dt, *J* = 10.0, 3.9 Hz, 1H), 2.32 (d, *J* = 11.4 Hz, 1H), 2.23 – 2.11 (m, 1H), 1.97 (dt, *J* = 33.3, 12.3 Hz, 6H), 1.77 – 1.53 (m, 9H), 1.43 (s, 9H), 1.36 – 1.01 (m, 7H), 1.06 – 0.88 (m, 13H), 0.86 (t, *J* = 7.2 Hz, 3H), 0.80 (t, *J* = 7.3 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{42}H_{69}N_4O_7]^+$ ($[M+H]^+$) 741.5161, found 741.5156.

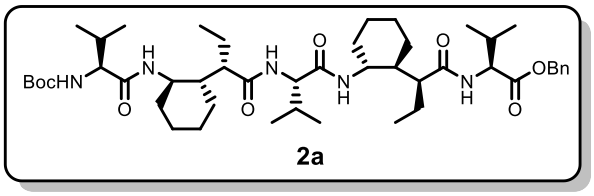
S8 – Boc-[^{L}Val]-OBn



S8

Boc-L-valine (1.09 g, 5.00 mmol) was dissolved to 0.1 M in DMF. To this solution was added Cs_2CO_3 (1 eq., 1.63 g, 5.00 mmol) and benzyl bromide (0.95 eq., 564 µL, 4.75 mmol). The reaction mixture was stirred for 12 hr at room temperature, after which it was washed with 0.5 M aqueous HCl, 5% aqueous LiCl, saturate aqueous $NaHCO_3$, and brine successively. The organic layer was dried over Na_2SO_4, filtered, and concentrated *in vacuo* to yield 1.78 g **S8** as a clear viscous oil which was used without further purification.

2a – Boc-([^{L}Val][(*S*,*S*,*R*)-EtACHA])$_2$[^{L}Val]-OBn

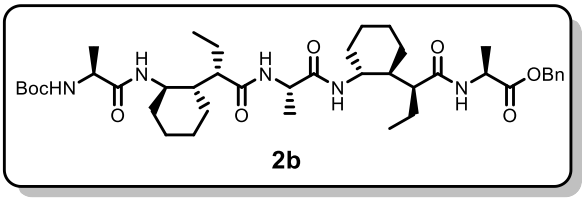


2a

Boc-L-valine benzyl ester **S8** (3 eq., 92 mg, 0.300 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, tetrapeptide **S7** (74 mg, 0.10 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.2 eq., 23.0 mg, 0.120 mmol), HOAt (1.2 eq., 200 µL from 0.6 M in DMF stock solution, 0.120 mmol), and DIEA (4 eq., 66 µL, 0.400 mmol) stirring at room temperature for 16 hr. Purification was carried out with silica column chromatography eluting with 2:1 hexanes/EtOAc to yield 33 mg pentapeptide **2a** as a white foam (0.39 mmol, 39% yield). **TLC** R_f = 0.49 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.93 (d, *J* = 8.6 Hz, 1H), 7.75 (d, *J* = 6.2 Hz, 1H), 7.41 – 7.29 (m, 5H), 6.76 (d, *J* = 9.1 Hz, 1H), 5.79 (d, *J* = 9.6 Hz, 1H), 5.24 (d, *J* = 12.3 Hz, 1H), 5.09 (d, *J* = 12.3 Hz, 1H), 4.97 (d, *J*

= 8.0 Hz, 1H), 4.61 (dd, *J* = 8.6, 5.4 Hz, 1H), 3.88 (t, *J* = 8.0 Hz, 1H), 3.74 – 3.58 (m, 3H), 2.32 (d, *J* = 11.0 Hz, 1H), 2.28 – 2.19 (m, 2H), 2.13 (dt, *J* = 11.8, 4.0 Hz, 1H), 2.10 – 2.03 (m, 1H), 1.93 (dddd, *J* = 23.2, 20.7, 11.7, 6.8 Hz, 5H), 1.74 – 1.56 (m, 6H), 1.43 (s, 9H), 1.35 – 1.06 (m, 10H), 1.03 – 0.93 (m, 16H), 0.91 (d, *J* = 6.9 Hz, 3H), 0.86 (t, *J* = 7.3 Hz, 3H), 0.76 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{47}H_{77}N_5NaO_8]^+$ ($[M+Na]^+$) 862.5665, found 862.5692.

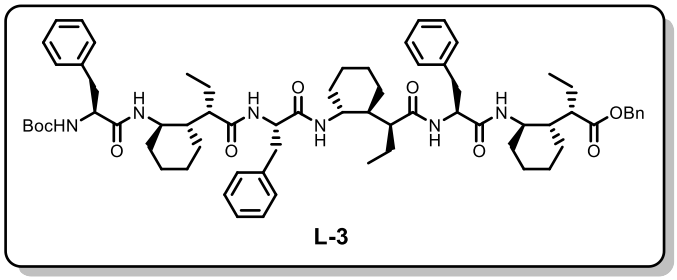
2b – Boc-([^{L}Ala][(*S*,*S*,*R*)-EtACHA])$_2$[^{L}Ala]-OBn



2b

Tetrapeptide *ent*-**1c** (1 eq., 83 mg, 0.12 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The deprotected acid was coupled according to **General Procedure 1** with L-alanine benzyl ester 4-toluenesulfonate salt (2 eq., 85 mg, 0.24 mmol), EDCI-HCl (2 eq., 46 mg, 0.24 mmol), HOBt (2 eq., 44 mg, 0.24 mmol), and DIEA (4 eq., 84 μL, 0.49 mmol) stirring at room temperature for 20 hr. Purification was carried out using automated silica column chromatography on a Biotage SNAP 10g cartridge eluting with a gradient of 50% → 60% EtOAc in hexanes over 10 column volumes to yield pentapeptide **2b** as 71 mg white solid (94 μmol, 78% yield). **TLC** R_f = 0.20 (5% MeOH in CH_2Cl_2, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.94 (d, *J* = 7.3 Hz, 1H), 7.68 (d, *J* = 5.5 Hz, 1H), 7.40 – 7.30 (m, 5H), 6.80 (d, *J* = 9.6 Hz, 1H), 5.95 (d, *J* = 9.6 Hz, 1H), 5.20 (d, *J* = 12.3 Hz, 1H), 5.11 (d, *J* = 12.3 Hz, 1H), 5.06 (d, *J* = 6.8 Hz, 1H), 4.67 (p, *J* = 7.2 Hz, 1H), 4.10 – 3.94 (m, 2H), 3.68 (qd, *J* = 10.9, 9.9, 3.1 Hz, 1H), 3.61 (qd, *J* = 10.9, 4.1 Hz, 1H), 2.19 (tt, *J* = 14.0, 3.4 Hz, 2H), 2.04 – 1.98 (m, 1H), 1.97 – 1.83 (m, 2H), 1.75 – 1.43 (m, 15H), 1.41 (s, 9H), 1.36 – 0.98 (m, 13H), 0.84 (t, *J* = 7.3 Hz, 3H), 0.75 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{41}H_{66}N_5O_8]^+$ ($[M+H]^+$) 756.4906, found 756.4905.

L-3 – Boc-([^{L}Phe][(*S*,*S*,*R*)-EtACHA])$_3$-OBn

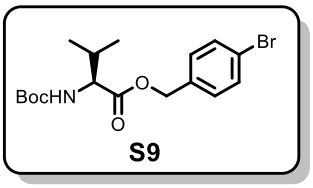


L-3

Boc-, benzyl ester-protected tetrapeptide **L-1a** (100 mg, 0.12 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, an amount of dipeptide **L-S3** (1.1 eq., 69 mg, 0.13 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.1 eq., 26 mg, 0.13 mmol), HOAt (1.1 eq., 223 μL from 0.6 M in DMF stock solution, 0.13 mmol), and DIEA (3 eq., 62 μL, 0.36 mmol) stirring at room temperature for 16 hr. Purification was carried

out with silica column chromatography eluting with 5:3 hexanes/EtOAc to yield **L-3** as 77 mg white foam (0.067 mmol, 56% yield). **TLC** R_f = 0.56 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 8.13 (d, *J* = 7.5 Hz, 1H), 7.73 (d, *J* = 5.5 Hz, 1H), 7.40 – 7.10 (m, 20H), 6.76 (d, *J* = 9.0 Hz, 1H), 6.29 (d, *J* = 9.7 Hz, 1H), 5.81 (d, *J* = 9.6 Hz, 1H), 5.14 (d, *J* = 7.2 Hz, 1H), 5.07 (d, *J* = 12.5 Hz, 1H), 5.03 (d, *J* = 12.5 Hz, 1H), 4.48 (q, *J* = 7.6 Hz, 1H), 4.37 (q, *J* = 7.6 Hz, 1H), 4.28 (dt, *J* = 9.7, 6.3 Hz, 1H), 3.64 (ddt, *J* = 14.4, 10.4, 5.3 Hz, 1H), 3.60 – 3.45 (m, 2H), 3.17 – 2.86 (m, 6H), 2.46 (dt, *J* = 10.2, 3.8 Hz, 1H), 2.18 (d, *J* = 10.1 Hz, 1H), 2.12 (d, *J* = 11.6 Hz, 1H), 1.99 (d, *J* = 12.7 Hz, 1H), 1.79 – 1.43 (m, 19H), 1.39 (s, 9H), 1.29 – 0.81 (m, 13H), 0.79 (t, *J* = 7.2 Hz, 3H), 0.71 (t, *J* = 7.2 Hz, 3H), 0.69 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{69}H_{95}N_6O_9]^+$ ($[M+H]^+$) 1151.7156, found 1151.7157.

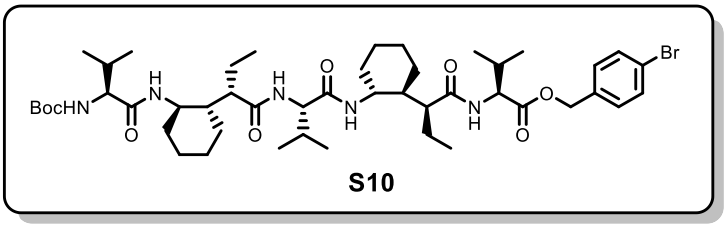
S9 − Boc-[^{L}Val]-OBrBn



S9

Boc-L-valine (652 mg, 3.00 mmol) was dissolved to 0.1 M in DMF. To this solution was added Cs_2CO_3 (1 eq., 977 mg, 3.00 mmol) and 4-bromobenzyl bromide (0.95 eq., 712 mg, 2.85 mmol). The reaction mixture was stirred for 12 hr at room temperature, after which it was washed with 0.5 M aqueous HCl, 5% aqueous LiCl, saturated aqueous $NaHCO_3$, and brine successively. The organic layer was dried over Na_2SO_4, filtered, and concentrated *in vacuo* to yield crude **S9** as 1.12 g clear viscous oil. This residue was filtered through a silica plug eluting with 4:1 hexanes/EtOAc to yield 1.04 g **S9** as a clear viscous oil (0.269 mmol, 90% yield). **TLC** R_f = 0.44 (4:1 hexanes/EtOAc, I_2 stain). **^{1}H NMR** (300 MHz, $CDCl_3$) δ 7.49 (d, *J* = 8.4 Hz, 2H), 7.23 (d, *J* = 8.3 Hz, 2H), 5.15 (d, *J* = 12.5 Hz, 1H), 5.07 (d, *J* = 12.5 Hz, 1H), 5.00 (d, *J* = 9.3 Hz, 1H), 4.26 (dd, *J* = 9.2, 4.7 Hz, 1H), 2.21 – 2.07 (m, 1H), 1.44 (s, 9H), 0.94 (d, *J* = 6.8 Hz, 3H), 0.85 (d, *J* = 6.9 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{17}H_{24}BrNO_4]^+$ ($[M+H]^+$) 386.0962, found 386.0949.

S10 − Boc-([^{L}Val][(*S*,*S*,*R*)-EtACHA])$_2$[^{L}Val]-OBrBn

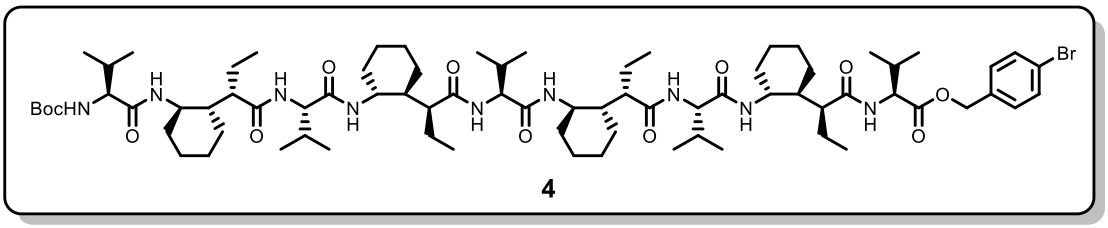


S10

Boc-L-valine *para*-bromobenzyl ester **X75** (3 eq., 156 mg, 0.405 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, **X56** (100 mg, 0.135 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (2 eq., 52 mg, 0.27 mmol), HOAt (2 eq., 450 μL from 0.6 M in DMF stock solution, 0.27 mmol), and DIEA (6 eq., 141 μL,

0.810 mmol) stirring at room temperature for 16 hr. Purification was carried out with silica column chromatography eluting with 2:1 hexanes/EtOAc to yield 75.9 mg white foam (0.826 mmol, 61% yield). **TLC** R_f = 0.48 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (300 MHz, $CDCl_3$) δ 7.94 (d, *J* = 8.5 Hz, 1H), 7.70 (d, *J* = 6.2 Hz, 1H), 7.50 (s, 1H), 7.25 (d, *J* = 8.6 Hz, 1H), 6.90 (d, *J* = 9.2 Hz, 1H), 6.02 (d, *J* = 9.7 Hz, 1H), 5.19 (d, *J* = 12.5 Hz, 1H), 5.14 (d, *J* = 7.2 Hz, 1H), 5.05 (d, *J* = 12.5 Hz, 1H), 4.57 (dd, *J* = 8.3, 5.6 Hz, 1H), 3.85 (t, *J* = 8.1 Hz, 1H), 3.75 – 3.61 (m, 3H), 2.53 – 2.09 (m, 4H), 2.04 – 1.53 (m, 10H), 1.42 (s, 9H), 1.37 – 0.80 (m, 36H), 0.76 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{47}H_{77}BrN_5O_8]^+$ ($[M+H]^+$) 918.4951, found 918.4947.

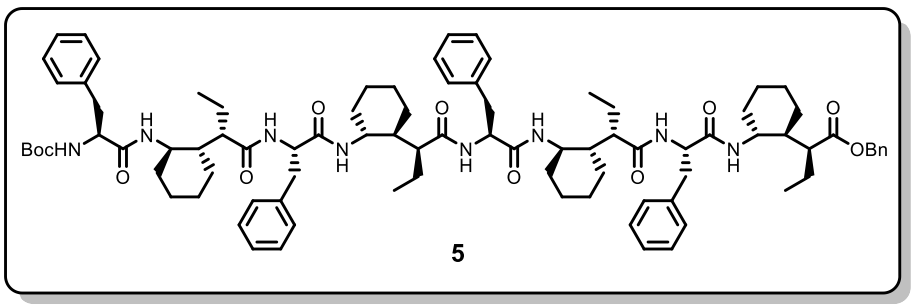
4 − Boc-([^{L}Val][(*S*,*S*,*R*)-EtACHA])$_4$[^{L}Val]-OBrBn



4

Boc-, benzyl ester-protected pentapeptide **S10** (33 mg, 0.035 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, an amount of tetrapeptide **S7** (1.1 eq., 29 mg, 0.039 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.1 eq., 7.5 mg, 0.039 mmol), HOAt (1.1 eq., 65 µL from 0.6 M in DMF stock solution, 0.039 mmol), and DIEA (3 eq., 19 µL, 0.11 mmol) stirring at room temperature for 40 hr. Purification was carried out with silica column chromatography eluting with 5:4 hexanes/EtOAc to yield 20 mg white foam (0.014 mmol, 39% yield). **TLC** R_f = 0.37 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 8.54 – 8.45 (m, 1H), 7.88 (d, *J* = 8.4 Hz, 1H), 7.75 (s, 1H), 7.49 (d, *J* = 8.3 Hz, 1H), 7.25 (d, *J* = 10.4 Hz, 1H), 6.98 (d, *J* = 9.2 Hz, 1H), 6.78 (d, *J* = 9.2 Hz, 1H), 6.67 (d, *J* = 9.3 Hz, 1H), 5.81 (d, *J* = 9.5 Hz, 1H), 5.19 (d, *J* = 12.5 Hz, 1H), 5.04 (d, *J* = 12.5 Hz, 1H), 4.96 (d, *J* = 7.9 Hz, 1H), 4.58 (dd, *J* = 8.4, 5.4 Hz, 1H), 3.96 (dd, *J* = 9.1, 6.1 Hz, 1H), 3.92 – 3.80 (m, 2H), 3.71 – 3.59 (m, 4H), 2.41 – 2.34 (m, 1H), 2.31 – 2.16 (m, 2H), 2.13 (dt, *J* = 12.0, 4.1 Hz, 1H), 2.09 – 1.80 (m, 12H), 1.75 – 1.48 (m, 19H), 1.43 (s, 9H), 1.36 – 0.90 (m, 52H), 0.87 – 0.74 (m, 12H). **MS** *m/z* (MALDI): calc. for $[C_{77}H_{128}BrN_8NaO_{11}]^+$ ($[M+Na]^+$) 1474.2, found 1474.2. Observed isotope pattern matches calculated distribution.

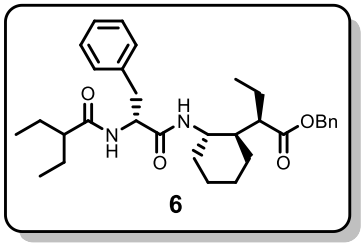
5 – Boc-([^{L}Phe][(*S*,*S*,*R*)-EtACHA])$_4$-OBn



5

Boc-, benzyl ester-protected tetrapeptide **L-1a** (53 mg, 64 μmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, another amount of tetrapeptide **L-1a** (1 eq., 53 mg, 64 μmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1 eq., 12 mg, 64 μmol), Oxyma Pure (1 eq., 9 mg, 64 μmol), and DIEA (2 eq., 22 μL, 128 μmol) stirring at room temperature for 36 hr. Purification was carried out with silica column chromatography eluting with 3% MeOH in CH_2Cl_2 to yield octapeptide **5** as 47 mg white foam (32 μmol, 50% yield). **TLC** R_f = 0.24 (5% MeOH in CH_2Cl_2, PMA stain). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.55 (d, *J* = 5.4 Hz, 1H), 8.22 (d, *J* = 7.6 Hz, 1H), 7.79 (d, *J* = 5.4 Hz, 1H), 7.40 – 7.10 (m, 25H), 6.66 (s, 1H), 6.38 (s, 1H), 6.14 (d, *J* = 9.6 Hz, 1H), 5.95 (d, *J* = 9.5 Hz, 1H), 5.17 – 4.94 (m, 4H), 4.51 (p, *J* = 7.2, 6.8 Hz, 3H), 4.20 (q, *J* = 7.4 Hz, 1H), 3.79 – 3.44 (m, 3H), 3.19 (dd, *J* = 14.5, 6.9 Hz, 1H), 3.13 – 2.82 (m, 7H), 2.52 – 2.40 (m, 2H), 1.82 – 1.45 (m, 20H), 1.39 (s, 9H), 1.31 – 0.72 (m, 29H), 0.73 – 0.61 (m, 9H). **HRMS** *m/z* (ESI): calc. for $[C_{89}H_{126}N_8O_{11}]^{2+}$ ($[M+2H]^{2+}$) 733.4611, found 733.4630.

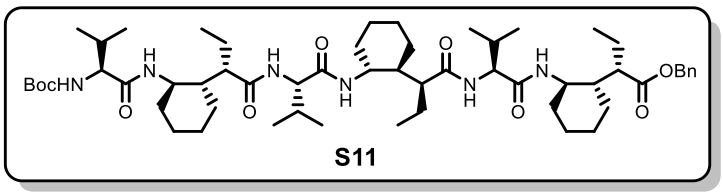
6 – $(CH_3CH_2)_2$CHCO-[^{D}Phe][(*R,R,S*)-EtACHA]-OBn



6

Boc-, benzyl ester-protected dipeptide **D-S3** (40 mg, 77 μmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Boc-deprotected **D-S3** was coupled to 2-ethylbutyric acid (1.5 eq., 12 μL, 92 μmol) according to **General Procedure 1** with EDCI-HCl (1.5 eq., 17 mg, 92 μmol), HOBt (1.5 eq., 21 mg, 92 μmol), and DIEA (3 eq., 32 μL, 0.18 mmol), stirring for 5 hr. Crude product was purified by automated silica column chromatography on a Biotage ZIP 10g cartridge using a gradient of 8% → 64% EtOAc in hexanes over 10 column volumes to yield **6** as 33 mg white foam (63 μmol, 82% yield). **TLC** R_f = 0.38 (2:1 hexanes:EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.38 – 7.18 (m, 10H), 6.21 (d, *J* = 7.7 Hz, 1H), 5.38 (d, *J* = 9.6 Hz, 1H), 5.08 (s, 2H), 4.59 (td, *J* = 8.3, 6.3 Hz, 1H), 3.66 (qd, *J* = 10.7, 3.7 Hz, 1H), 3.11 (dd, *J* = 13.5, 6.2 Hz, 1H), 2.94 (dd, *J* = 13.5, 8.7 Hz, 1H), 2.32 (dt, *J* = 11.1, 3.2 Hz, 1H), 1.86 (tt, *J* = 9.3, 5.2 Hz, 1H), 1.69 – 1.33 (m, 12H), 1.27 – 0.94 (m, 3H), 0.79 (q, *J* = 7.5 Hz, 9H). **HRMS** *m/z* (ESI): calc. for $[C_{32}H_{45}N_2O_4]^+$ ($[M+H]^+$) 521.3374, found 521.3372.

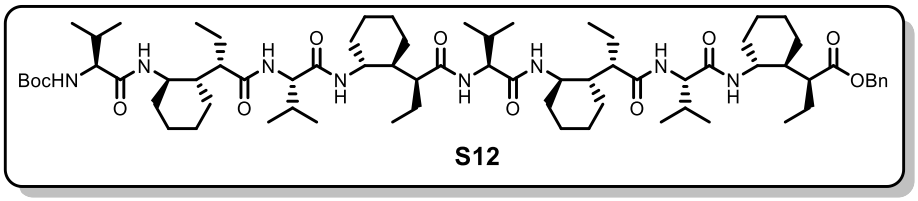
S11 – Boc-([^{L}Val][(*S,S,R*)-EtACHA])$_3$-OBn



S11

Boc-, benzyl ester-protected tetrapeptide **S7** (90 mg, 0.12 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, an amount of dipeptide **S6** (1.1 eq., 63 mg, 0.13 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.1 eq., 26 mg, 0.13 mmol), HOAt (1.1 eq., 223 μL from 0.6 M in DMF stock solution, 0.13 mmol), and DIEA (2 eq., 43 μL, 0.24 mmol) stirring at room temperature for 40 hr. Purification was carried out with silica column chromatography eluting with 3:2 hexanes/EtOAc to yield hexapeptide **S11** as 54 mg white foam (0.054 mmol, 44% yield). **TLC** R_f = 0.37 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.01 (d, *J* = 6.1 Hz, 1H), 7.82 (d, *J* = 8.5 Hz, 1H), 7.33 (q, *J* = 7.1, 6.1 Hz, 5H), 6.85 (d, *J* = 9.0 Hz, 1H), 6.69 (d, *J* = 9.7 Hz, 1H), 6.07 (d, *J* = 9.0 Hz, 1H), 5.17 (d, *J* = 12.5 Hz, 1H), 5.13 (d, *J* = 12.3 Hz, 1H), 4.93 (d, *J* = 7.9 Hz, 1H), 4.32 (dd, *J* = 8.6, 5.9 Hz, 1H), 3.99 (t, *J* = 7.9 Hz, 1H), 3.84 – 3.56 (m, 4H), 2.54 (d, *J* = 10.9 Hz, 1H), 2.47 – 2.40 (m, 1H), 2.39 – 2.29 (m, 1H), 2.13 – 1.45 (m, 22H), 1.42 (s, 9H), 1.38 – 0.92 (m, 32H), 0.89 (t, *J* = 7.3 Hz, 3H), 0.82 (t, *J* = 7.1 Hz, 3H), 0.72 (t, *J* = 7.2 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{57}H_{94}N_6NaO_9]^+$ ($[M+Na]^+$) 1029.6975, found 1029.6953.

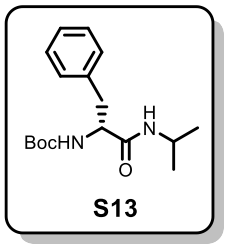
S12 – Boc-([^{L}Val][(*S,S,R*)-EtACHA])$_4$-OBn



S12

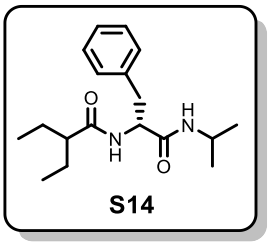
Boc-, benzyl ester-protected tetrapeptide **S7** (93 mg, 0.13 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. Separately, another amount of tetrapeptide **S7** (1.2 eq., 111 mg, 0.15 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. The two components were coupled according to **General Procedure 1** with HATU (1.2 eq., 57 mg, 0.15 mmol), HOAt (1.2 eq., 250 μL from 0.6 M in DMF stock solution, 0.15 mmol), and DIEA (2 eq., 44 μL, 0.25 mmol) stirring at room temperature for 48 hr. Purification was carried out with silica column chromatography eluting with 4:3 hexanes/EtOAc to yield octapeptide **S12** as 72 mg white foam (0.048 mmol, 45% yield). **TLC** R_f = 0.39 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.64 (d, *J* = 5.6 Hz, 1H), 7.81 (d, *J* = 8.6 Hz, 1H), 7.73 (d, *J* = 6.1 Hz, 1H), 7.41 – 7.29 (m, 5H), 7.07 (d, *J* = 9.3 Hz, 1H), 6.81 (d, *J* = 9.1 Hz, 1H), 6.71 (d, *J* = 9.7 Hz, 1H), 6.10 (d, *J* = 9.1 Hz, 1H), 5.16 (d, *J* = 12.6 Hz, 1H), 5.13 (d, *J* = 12.6 Hz, 1H), 4.97 (d, *J* = 8.0 Hz, 1H), 4.32 (dd, *J* = 8.5, 5.8 Hz, 1H), 4.05 (dd, *J* = 9.1, 6.1 Hz, 1H), 3.85 (t, *J* = 7.9 Hz, 1H), 3.81 – 3.52 (m, 4H), 3.44 – 3.33 (m, 1H), 2.55 (dt, *J* = 10.9, 3.5 Hz, 1H), 2.48 (d, *J* = 7.9 Hz, 1H), 2.40 – 2.26 (m, 2H), 2.24 – 2.09 (m, 3H), 2.09 – 1.56 (m, 24H), 1.43 (s, 9H), 1.38 – 0.72 (m, 57H). **HRMS** *m/z* (ESI): calc. for $[C_{72}H_{121}N_8O_{11}]^+$ ($[M+H]^+$) 1273.9150, found 1273.9127.

S13 – Boc-[^{D}Phe]-NH^{i}Pr



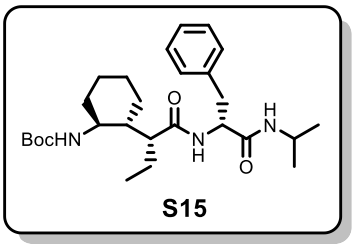
Boc-D-phenylalanine (1 eq., 571 mg, 2.00 mmol) was coupled to isopropylamine (1.5 eq., 379 µL, 3.00 mmol) according to **General Procedure 1** with EDCI-HCl (1.5 eq., 575 mg, 3.00 mmol), HOBt (1.5 eq., 541 mg, 3.00 mmol), and DIEA (3 eq., 1.05 mL, 6.00 mmol) and stirred for 2.5 hr. Crude product was purified with silica column chromatography eluting with 2:1 hexanes:EtOAc to yield **S13** as 323 mg white solid (0.62 mmol, 31%). **TLC** R_f = 0.50 (1:1 hexanes:EtOAc, ninhydrin stain) **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.29 – 7.14 (m, 5H), 6.49 (d, *J* = 8.0 Hz, 1H), 5.68 (d, *J* = 8.5 Hz, 1H), 4.41 (q, *J* = 7.7 Hz, 1H), 4.02 – 3.88 (m, 1H), 3.00 (d, *J* = 7.4 Hz, 2H), 1.39 (s, 9H), 1.04 (d, *J* = 6.7 Hz, 3H), 0.96 (d, *J* = 6.6 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{17}H_{27}N_2O_3]^+$ ($[M+H]^+$) 307.2017, found 307.2017.

S14 – $(CH_3CH_2)_2$CHCO-[^{D}Phe]-NH^{i}Pr



Boc-protected **X1** (1 eq., 59 mg, 0.19 mmol) was Boc-deprotected according to **General Procedure 2**. The product of this reaction were coupled with 2-ethylbutyric acid (1.2 eq., 29 µL, 0.23 mmol) according to **General Procedure 1** with EDCI-HCl (1.2 eq., 44 mg, 0.23 mmol), DMAP (0.2 eq., 5 mg, 40 µmol), and DIEA (2.4 eq., 80 µL, 0.461 mmol) and stirred for 16 hr. Product **S14** was isolated as 36 mg white solid (0.12 mmol, 61% yield) after extractive workup, which was highly pure and did not require silica column chromatography. **TLC** R_f = 0.40 (1:1 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.23 – 7.11 (m, 5H), 6.22 (d, *J* = 7.9 Hz, 1H), 5.73 (d, *J* = 7.9 Hz, 1H), 4.57 (q, *J* = 7.6 Hz, 1H), 3.88 (h, *J* = 6.7 Hz, 1H), 3.03 (dd, *J* = 13.7, 7.0 Hz, 1H), 2.93 (dd, *J* = 13.7, 7.9 Hz, 1H), 1.81 (tt, *J* = 9.4, 5.1 Hz, 1H), 1.48 (tp, *J* = 15.5, 7.7 Hz, 2H), 1.42 – 1.28 (m, 2H), 0.97 (d, *J* = 6.6 Hz, 3H), 0.91 (d, *J* = 6.5 Hz, 3H), 0.76 (t, *J* = 7.4 Hz, 3H), 0.68 (t, *J* = 7.4 Hz, 3H). **HRMS** *m/z* (ESI): calc. for $[C_{18}H_{29}N_2O_2]^+$ ($[M+H]^+$) 305.2224, found 305.2220.

S15 – Boc-[(*R*,*R*,*S*)-EtACHA][^{D}Phe]-NH^{i}Pr



D-S2 (1 eq., 46 mg, 0.12 mmol) was benzyl-deprotected at the C-terminus according to **General Procedure 3**. Boc-protected **X1** (1.2 eq., 45 mg, 0.15 mmol) was Boc-deprotected at the N-terminus according to **General Procedure 2**. The two components were coupled according to **General Procedure 1** with EDCI-HCl (1.2 eq., 28 mg, 0.15 mmol), Oxyma Pure (1.2 eq., 20 mg, 0.15 mmol), and DIEA (2.4 eq., 52 µL, 0.29 mmol) stirring at room temperature for 20 hr. Crude product was purified with silica column chromatography eluting with 3:2 hexanes:EtOAc to yield **S15** as 48 mg white solid (0.10 mmol, 82% yield). **TLC** R_f = 0.15 (1:2 hexanes/EtOAc, PMA stain). **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.35 – 7.19 (m, 5H), 6.38 (d, *J* = 7.6 Hz, 1H), 5.69 (d, *J* = 7.9 Hz, 1H), 4.58 (q, *J* = 7.6 Hz, 1H), 4.50 (d, *J* = 9.7 Hz, 1H), 3.97 (h, *J* = 6.7 Hz, 1H), 3.35 (qd, *J* = 10.8, 3.8 Hz, 1H), 3.14 (dd, *J* = 13.7, 7.0 Hz, 1H), 3.00 (dd, *J* = 13.7, 8.1 Hz, 1H), 2.32 – 2.24 (m, 1H), 2.00 – 1.91 (m, 1H), 1.73 – 1.52 (m, 6H), 1.45 (s, 9H), 1.31 – 1.07 (m, 4H), 1.05 (d, *J* = 6.7 Hz, 3H), 0.98 (d, *J* = 6.6 Hz, 3H), 0.79 (t, *J* = 7.2 Hz, 3H). Small peaks (integration ≈ 0.11) were identified as rotamers with gradient 1D-NOESY with 2 s mixing time.[S2] **HRMS** *m/z* (ESI): calc. for $[C_{27}H_{44}N_3O_4]^+$ ($[M+H]^+$) 474.3330, found 474.3327.

III. Tetramer Crystal Structures

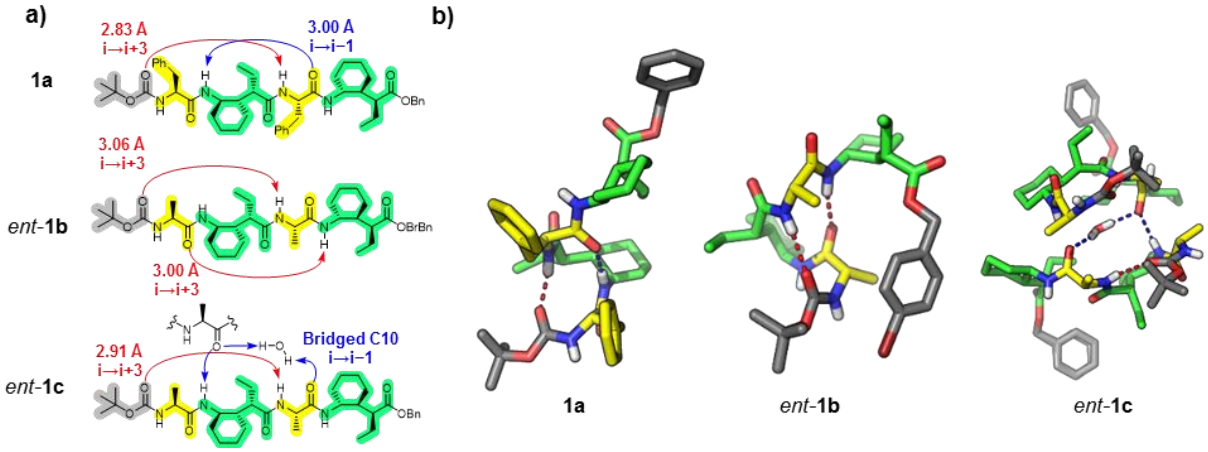


Figure S1. a) Structures of tetrapeptides **1a-c** depicting H-bonds observed in the crystal structures. Distances shown are acceptor-donor atomic distances A−D where A⟶H−D is the hydrogen bond. **b)** Crystal structures of tetrapeptides **1a-c**. Enantiomers of **1b** and **1c** are shown for stereochemical consistency with the other crystal structures.

IV. 2D-NMR

a. 2D-NMR Experimental

Peptides **2a**, **4**, **5**, **S12**, and **S11** were prepared for 2D-NMR analysis by dissolving to 4-10 mM in 500-600 µL $CDCl_3$ (0.03% TMS) dried over 3Å molecular sieves and transferred to 5 mm Norell Sample Vault NMR tubes (SVCP-5-178). Prior to 2D-NMR data collection, good peptide solubility was confirmed by measuring the ^{1}H-NMR at concentrations of 100 µM to 10 mM; $\Delta\delta$ of amide NH < 0.02 ppm was interpreted as lack of aggregation.

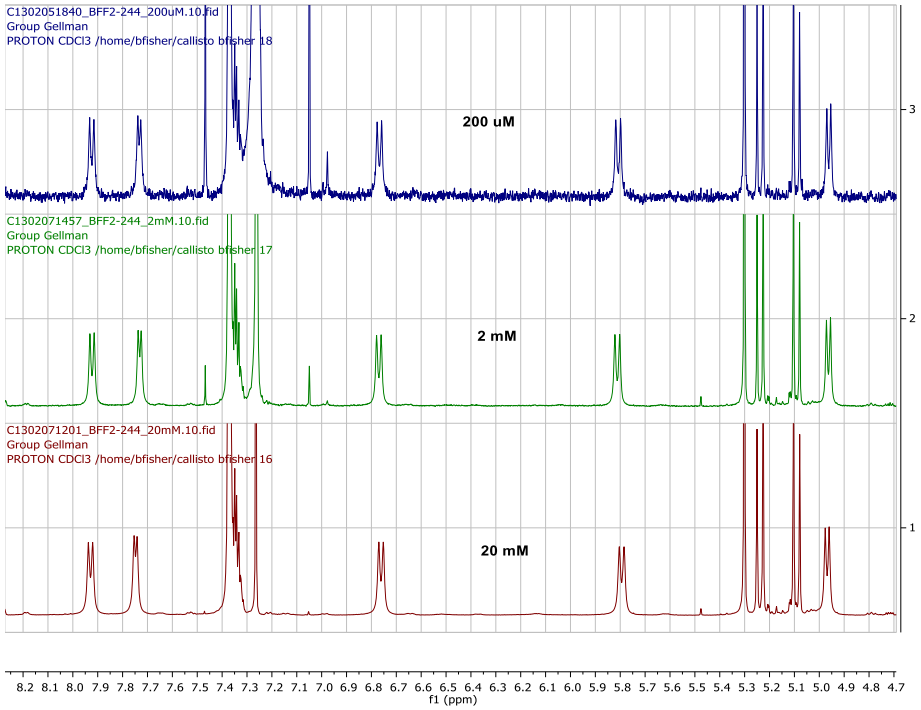


Figure S2. Dilution studies of **2a**. Change in chemical shift $\Delta\delta$ < 0.02 for all amide/carbamate NH from 200 µM to 20 mM, interpreted as lack of significant aggregation under 2D NMR experimental conditions.

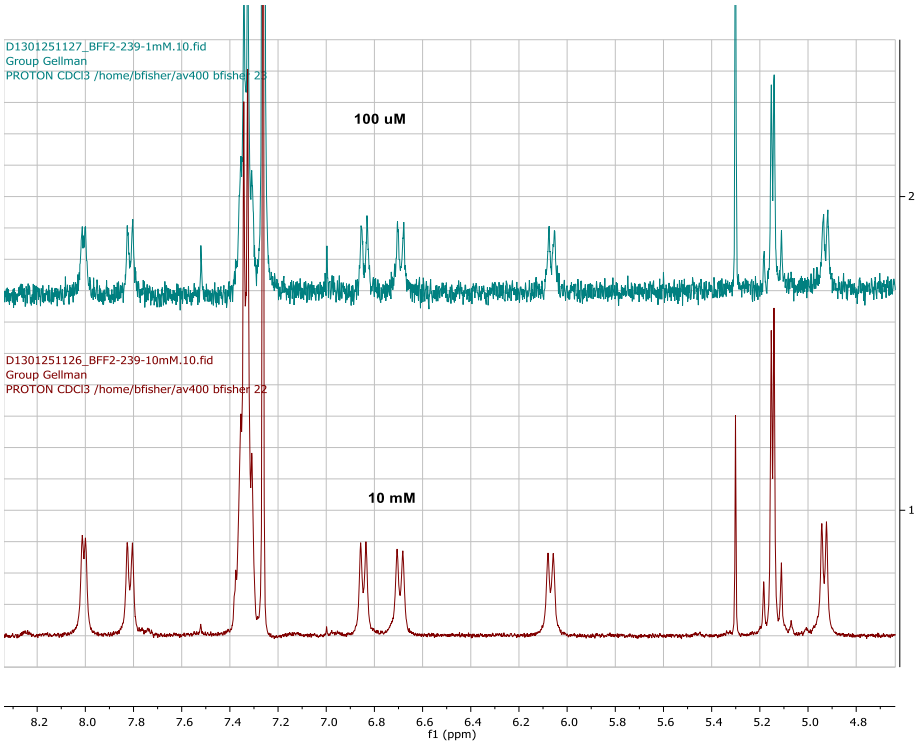


Figure S3. Dilution studies of **S11**. Change in chemical shift $\Delta\delta < 0.02$ for all amide/carbamate NH from 100 μM to 10 mM, interpreted as lack of significant aggregation under 2D NMR experimental conditions

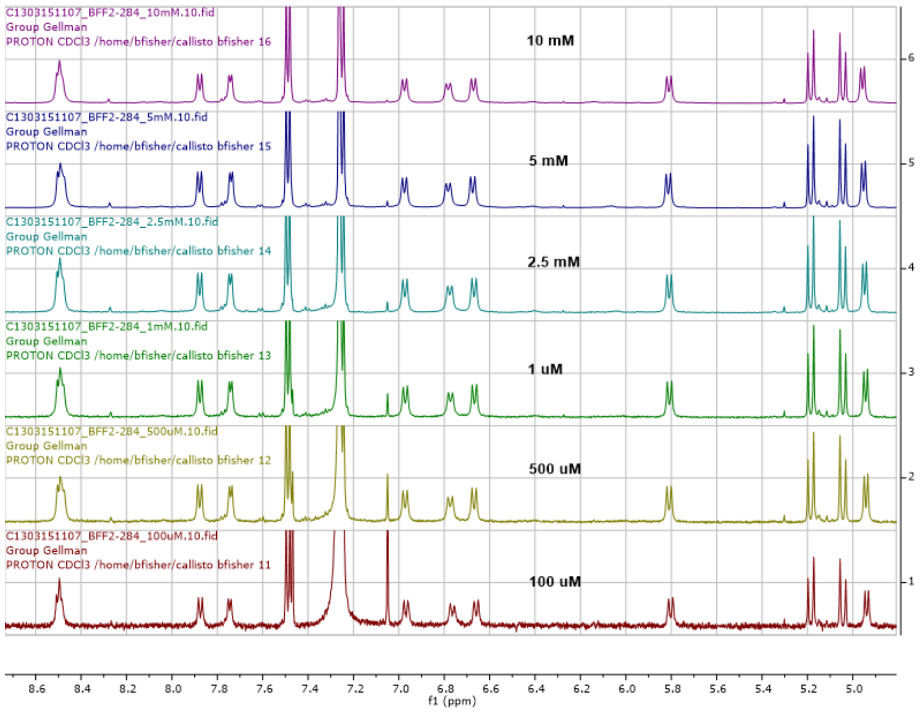


Figure S4. Dilution studies of **4**. Change in chemical shift $\Delta\delta < 0.02$ for all amide/carbamate NH from 100 μM to 10 mM, interpreted as lack of significant aggregation under 2D NMR experimental conditions.

2D-NMR data of compounds **2a**, **4**, **5**, and **S11** were collected at 24°C on a Bruker 500 MHz Avance III-500 spectrometer equipped with a DCH cryoprobe and running in Icon mode. Most parameters were unchanged from their default values; parameters altered are shown in **Table S1**.[S3] Sweep width was manually optimized from the ^{1}H-NMR data.

Table S1. Selected parameters in 2D-NMR experiments of **2a**, **S11**, **4**, and **5**.

Name	2a	S11	4	5
Instrument	Bruker 500 MHz	Bruker 500 MHz	Bruker 500 MHz	Bruker 500 MHz
Concentration	10 mM	10 mM	10 mM	10 mM
COSY parameter set	COSYGPSW	COSYGPSW	COSYGPSW	COSYGPSW
COSY pulseprog	cosygpppqf	cosygpppqf	cosygpppqf	cosygpppqf
ns	4	2	4	4
ds	8	8	8	8
td(f2)	2048	2048	2048	2048
td(f1)	256	256	256	256
TOCSY parameter set	MLEVPHSW	MLEVPHSW	MLEVPHSW	MLEVPHSW
TOCSY pulseprog	mlevphpp	mlevphpp	mlevphpp	mlevphpp
ns	8	8	8	4
ds	16	16	16	16
td(f2)	2048	2048	2048	2048
td(f1)	256	256	256	256
D9	80 ms	80 ms	80 ms	80 ms
NOESY/ROESY parameter set	NOESYPHSW	NOESYPHSW	ROESYPHSW	ROESYPHSW
NOESY/ROESY pulseprog	noesygpphpp	noesygpphpp	roesyphpp.2	roesyphpp.2
ns	16	16	16	16
ds	32	32	32	32
td(f2)	2048	2048	2048	2048
td(f1)	256	256	256	256
p15	–	–	200 ms	200 ms
d8	300 ms	300 ms	–	–

2D-NMR data of **2a**, **S11**, **4**, and **5** were processed with Bruker TopSpin 3.1 initially using default processing parameters defined in the parameter sets. COSY data were baseline fit with default settings. TOCSY and ROESY data were phased in both dimensions automatically and adjusted manually, then baseline fit in both dimensions with default settings. The computer program SPARKY[S4] was used for resonance assignment and analysis of the processed data. ROESY crosspeak volume integration was carried out using default parameters and inspected for accuracy.

2D-NMR data (gCOSY, TOCSY, ROESY) of 4 mM compound **S12** in dry $CDCl_3$ was collected on a Varian INOVA-600 MHz spectrometer equipped with a 5 mm probe at 10°C. Standard Varian pulse sequences were used and the data were processed using Varian VNMR 6.1 software. Shifted sine bell window functions were generally applied before Fourier transformation. The gCOSY spectrum was obtained in absolute mode with gradient echo coherence selection; TOCSY and ROESY spectra were acquired in the sensitive mode with hypercomplex phase cycling (States-Haberkorn method). All experiments were performed by collecting 2048 points in f2 and 300-600 points in f1. The TOCSY experiment employed a standard MLEV-17 spin lock sequence with a

spin lock field of 7-8 KHz and mixing time of 80 ms. The ROESY experiment used spin-locking fields of ~3 kHz and mixing time of 250 ms. The computer program SPARKY[S4] was used for resonance assignment and analysis of the processed data. ROESY crosspeak volume integration was carried out using default parameters and inspected for accuracy.

b. 2D-NMR Resonance/ROESY Distance Assignment Procedures

Sequential Assignment Procedure

Resonance assignment of peptide foldamers was accomplished by employing sequential resonance assignment from the N-terminus to the C-terminus.[S5] The N-terminal Boc carbamate NH ($\delta \approx 5.0$ ppm) is invariably upfield of all amide NHs of a peptide ($\delta > 5.5$ ppm) and has a characteristic H-N broadness distinguishing it from H-C resonances that may be nearby. Thus, assignment of the Boc carbamate NH is nearly unambiguous, and assignment of the rest of the peptide's resonances can rely on relaying information from this proton through to the C-terminus of the peptide sequence. The carbamate NH allows for simple assignment of the rest of the protons in the spin system, H_α and sidechain protons of the first α-residue, using the COSY and TOCSY data. A very intense ROESY crosspeak is found between $H_\alpha(\alpha,i)$ and $HN(\gamma,i+1)$. Taking the expected chemical shift range of H_γ into consideration ($\delta \approx 3.4$-3.6), a single likely resonance assignment emerges for $HN(\gamma,i+1)$, extending assignment past the peptide bond. Assignment of the protons attached to functionalized carbons (the unfunctionalized cyclohexyl protons are too overlapped for unambiguous assignment) in the rest of the γ-residue spin system is readily achieved with the COSY and TOCSY data. A very intense ROESY crosspeak can be found between $H_\alpha(\gamma,i+1)$ and $HN(\alpha,i+2)$, allowing for its unambiguous assignment and leaping across another peptide bond. These steps are followed until full resonance assignment is achieved. Confirmation of the assignments can be achieved using HMBC to relate the unique ester carbonyl at the C-terminus to the H_α of the last residue.

NOE Distance Calculation

Integrated ROESY crosspeak volumes were converted into distances r_{ij} (assuming the initial rate approximation) by r^{-6} averaging.[S6] Comparison was made with an integrated known reference distance r_{ref} using **Equation 1**:

$$r_{ij} = r_{ref}\left(\frac{I_{ref}}{I_{ij}}\right)^{1/6} \quad \textbf{(1)}$$

Where I_{ij} represents the multiplicity-corrected volume integral of the crosspeak involving nuclei *i* and *j*. For crosspeaks involving only two protons, I_{ij} is simply equal to the measured crosspeak integral I_{meas}. For crosspeaks involving groups with multiple equivalent or stereo-unassigned protons, the measured integral is corrected using **Equation 2:**

$$I_{ij} = \frac{I_{meas}}{n_i \times n_j} \quad \textbf{(2)}$$

Where n_i is the number of nuclei *i* and n_j is the number of nuclei *j*.

Selection of reference distances

Two major factors must be considered in selecting a ROESY crosspeak to use as a reference distance. First, and most obviously, the internuclear distance between the two protons forming the

crosspeak must be known and fixed. Second, also obviously, the crosspeak must be well separated from other peaks in the spectrum to ensure accurate integration.

For pentamer **2a**, the diastereotopic methylene CH_2 protons are well-resolved from one another, and the methyl CH_3 of the ethyl sidechain of [γ(**I**)]4 are at a fixed distance of 2.40 Å from these protons. The crosspeak between one of these methylene protons and the CH_3 was used as the reference distance (after multiplicity correction by dividing the integral by 3).

Hexamer **S11** also featured well-resolved diastereotopic methylene protons on the ethyl sidechain of [γ(**I**)]4, and the 2.40 Å distance between one of these protons and the CH_3 methyl protons was used as the reference distance after multiplicity correction.

In octamer **S12**, the two diastereotopic methylene protons of the ethyl sidechain of γ(**I**) were found to be well resolved. The selected reference distance was the 2.40 Å fixed distance between one of these protons and the well-resolved methyl protons, with multiplicity 3, of the ethyl sidechain.

The design of nonamer **4** included 4-bromobenzyl ester as the C-terminal protecting group in order to incorporate well-resolved *ortho*-aryl protons, each of multiplicity 2, with a covalently-fixed internuclear distance of 2.80 Å. Zero-quantum artifacts made integration of these peaks difficult. The positive and negative phases were found to be approximately equivalent, and the absolute values of these were added to provide I_{ref} after division by the product of the number of protons involved in this crosspeak ($2 \times 2 = 4$ in this case).

The diastereotopic H_β of residue [Phe]3 in octamer **5**, fixed at 1.78 Å from one another, were sufficiently well resolved for them to be used as the fixed reference distance.

c. Model Helices

In order to provisionally assign non-sequential NOEs to a particular secondary structure, helical conformations of model α/γ(**I**)-peptide **S16** were constructed with several different H-bonding patterns (**Figure S4**). Observed non-sequential NOEs were compared to the expected NOEs of each model, and the model with the greatest number of expected NOEs observed experimentally was assigned as the likeliest candidate. Model α/γ(**I**)-peptide helices were constructed by using Avogadro software[S7] to build the appropriate sidechains of L-valine and (*S*,*S*,*R*)-EtACHA onto the .pdb files included in the Supporting Information in Hofmann *et al.*'s computational study of the unsubstituted α/γ-peptide backbone.[S8] The helical conformations investigated were the 12/10-helix, the 12-helix, and the 20/18-helix, since these were the lowest energy conformations found of the unsubstituted α/γ-peptide backbone by Hofmann that featured the α-residue in an allowed region of the Ramachandran plot. Other conformations, including the 10/12- and 18/20-helix, were excluded since they would require α-residues to adopt ϕ/ψ dihedral angles in forbidden regions of the Ramachandran plot. Molecular mechanics geometry optimization within Avogadro was performed prior to Gaussian 09[S9] geometry optimization at the B3LYP/6-31G level of theory using WebMO[S10] as the submission interface. PyMOL[S11] was used to calculate proton internuclear distances of the structures.

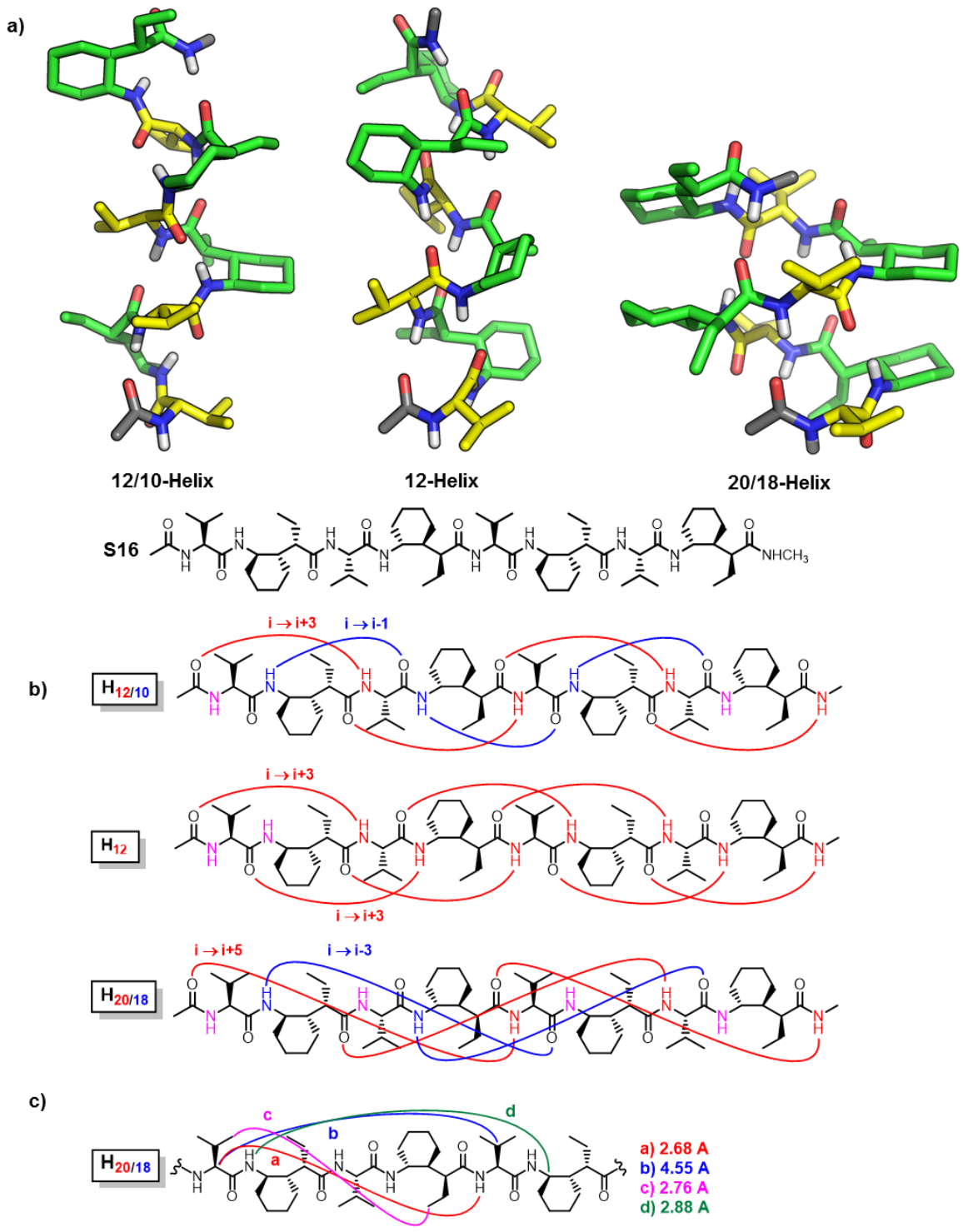
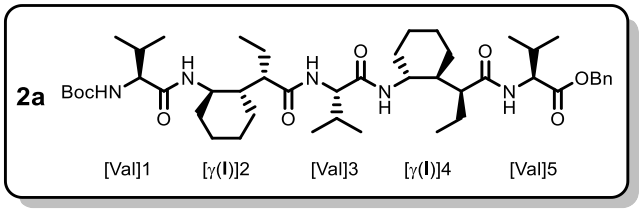


Figure S4. a) B3LYP/6-31G structures of the 12/10-, 12-, and 20/18-helical conformations of **S16**. Gray represents terminal acetyl and methylamide caps, yellow represents α-residues, and green represents γ-residues. **b)** H-bonding patterns in the helical conformations. **c)** Expected NOEs in the 20/18-helical conformation. None are observed in the ROESY spectra of **2a**, **S11**, **4**, **5**, or **S12**.

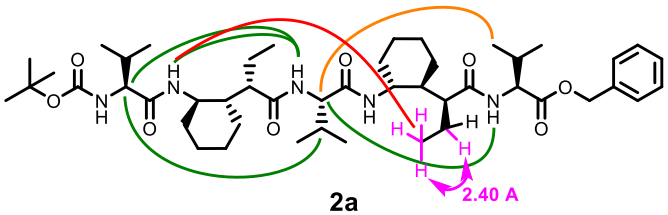
d. Resonance Assignments and ROESY-Derived Interproton Distances of 2a, S11, 4, 5, and S12

Table S2. Resonance assignment of **2a**



	Backbone				Sidechain	
Residue Number	**NH**	**γ**	**β**	**α**	**β2**	**γ3**
N-Terminus					Boc: 1.428	
[Val]1	4.965			3.875	1.884	0.987
[γ(I)]2	6.762	3.646		2.125	1.430, 1.498	0.765
[Val]3	7.743			3.631	1.968	0.993
[γ(I)]4	5.796	5.796		2.321	1.468, 1.640	0.860
[Val]5	7.926			4.612	2.231	0.909, 0.959
C-Terminus					Bn: 5.087, 5.226	Ph: 7.370

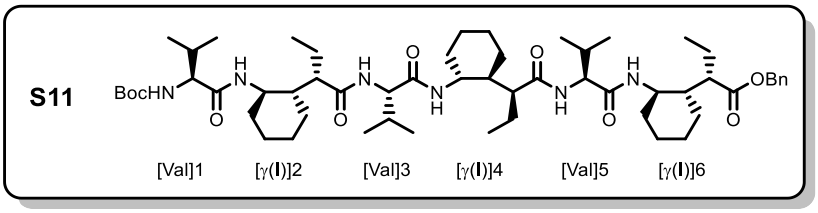
Table S3. ROESY crosspeaks of **2a**



Atom 1	Atom 2	Distance	Designation
Reference distance			
[γ(I)]4(Et12)	[γ(I)]4(Et2)	2.40	
Sequential NOEs			
[Val]1(Hα)	[γ(I)]2(HN)	2.0	Strong
[γ(I)]2(Et2)	[Val]3(HN)	3.1	Medium
[γ(I)]2(Hα)	[Val]3(HN)	1.9	Strong
[γ(I)]2(Et2)	[Val]3(Hα)	3.0	Medium
[Val]3(Hα)	[γ(I)]4(HN)	1.9	Strong
[Val]3(HG)	[γ(I)]4(HN)	3.3	Medium
[γ(I)]4(Hα)	[Val]5(HN)	1.9	Strong
[γ(I)]4(Et2)	[Val]5(HN)	3.7	Weak
Non-sequential NOEs			
[Val]1(Hα)	[Val]3(HB)	2.3	Strong
[Val]1(Hα)	[Val]3(HN)	2.4	Strong
[γ(I)]2(HN)	[γ(I)]4(Et2)	3.7	Weak
[γ(I)]2(HN)	[Val]3(HN)	2.3	Strong

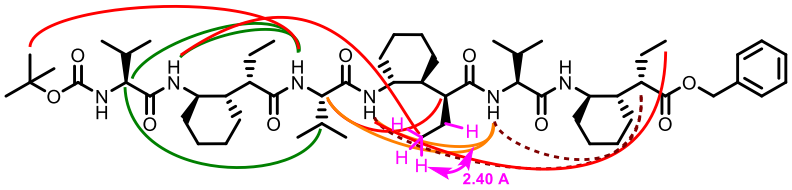
[Val]3(Hα)	[Val]5(Hγ1)	3.0	Medium
[Val]3(Hα)	[Val]5(HN)	2.4	Strong
[Val]3(Hα)	[Val]5(Ph)	3.6	Weak

Table S4. Resonance assignment of **S11**

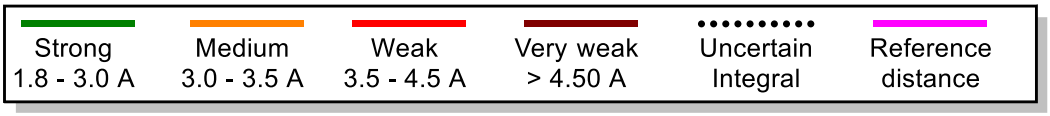


	Backbone				Sidechain	
Residue Number	**NH**	**γ**	**β**	**α**	**β2**	**γ3**
N-Terminus					Boc: 1.421	
[Val]1	4.935			3.989	1.869	1.002
[γ(I)]2	6.846	3.633	1.724	2.085		0.727
[Val]3	8.004			3.611	1.949	0.980
[γ(I)]4	6.698	3.727	1.955	2.446		0.894
[Val]5	7.816			4.321	2.342	0.978
[γ(I)]6	6.069	3.649	2.032	2.546		0.802
C-Terminus					Bn: 5.148	Ph: 7.331

Table S5. ROESY crosspeaks of **S11**



S11

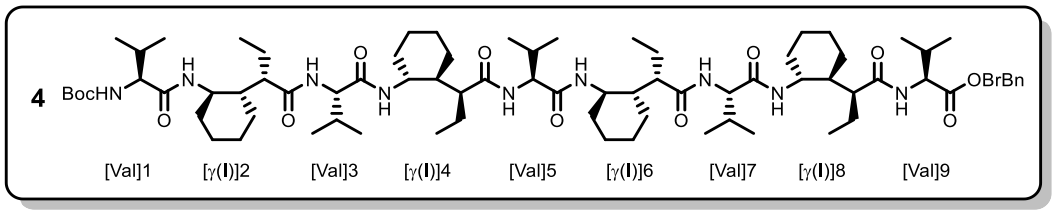


Atom 1	Atom2	Distance	Designation
Reference distance			
[γ(I)]4(Et12)	[γ(I)]4(Et2)	2.4	
Sequential NOEs			
[Val]1(Hα)	[γ(I)]2(HN)	1.9	Strong
[γ(I)]2(HN)	[γ(I)]2(Hα)	2.5	Strong
[γ(I)]2(Hα)	[Val]3(HN)	1.9	Strong
[γ(I)]2(Et2)	[Val]3(HN)	3.2	Medium
[γ(I)]2(HN)	[Val]3(HN)	2.5	Strong
[Val]3(Hα)	[γ(I)]4(Hα)	3.7	Weak
[Val]3(Hα)	[γ(I)]4(HN)	2.0	Strong
[γ(I)]4(Hα)	[Val]5(HN)	1.9	Strong
[γ(I)]4(Et2)	[Val]5(HN)	2.9	Strong
[γ(I)]4(Et2)	[Val]5(Hα)	3.7	Medium

[Val]5(Hα)	[γ(I)]6(HN)	2.3	Strong
[Val]5(Hβ)	[γ(I)]6(HN)	2.6	Strong
[Val]5(Hγ)	[γ(I)]6(HN)	3.4	Medium
[Val]5(HN)	[γ(I)]6(HN)	2.4	Strong
Non-sequential NOEs			
[Val]1(Boc)	[Val]3(HN)	4.0	Weak
[Val]1(Hα)	[Val]3(HN)	2.5	Strong
[Val]1(Hα)	[Val]3(Hβ)	2.4	Strong
[γ(I)]2(HN)	[γ(I)]4(Et2)	3.1	Medium
[Val]3(Hα)	[Val]5(HN)	3.1	Medium
[Val]3(Hα)	[γ(I)]6(Bn)	3.4	Medium
[γ(I)]4(HN)	[Val]5(HN)	3.3	Medium
[γ(I)]4(HN)	[γ(I)]6(Bn)	4.6	Weak
[γ(I)]4(HN)	[γ(I)]6(Hα)	≈ 4.5[a]	Very weak
[γ(I)]4(HN)	[γ(I)]6(Et2)	3.8	Weak
[Val]5(Hγ)	[γ(I)]6(Bn)	3.4	Medium
[Val]5(HN)	[γ(I)]6(Hα)	≈ 4.5[a]	Very weak

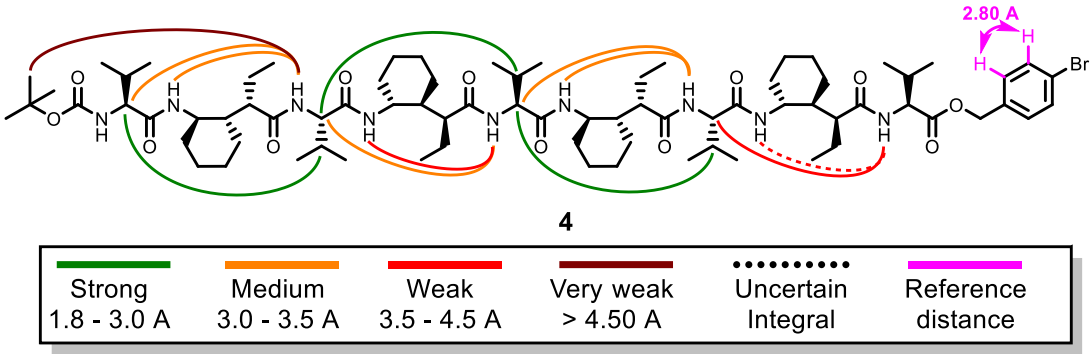
[a]Too weak to reliably integrate

Table S6. Resonance assignment of **4**



	Backbone				Sidechain	
Residue Number	**NH**	**γ**	**β**	**α**	**β2**	**γ3**
N-Terminus					Boc: 1.432	
[Val]1	4.958			3.871	1.905	0.988
[γ(I)]2	6.978	3.652		2.137		0.766
[Val]3	7.741			3.963	1.942	0.998
[γ(I)]4	6.786	3.647		2.165		0.817
[Val]5	8.487			3.836	1.888	0.971
[γ(I)]6	6.675	3.651		2.266		0.817
[Val]7	8.501			3.654	1.973	0.996
[γ(I)]8	5.811	3.657		2.376	1.480, 1.627	0.845
[Val]9	7.880			4.589	2.213	0.915; 0.955
C-Terminus					Bn: 5.044, 5.188	Ph1,2: 7.247, 7.500

Table S7. ROESY crosspeaks of **4**

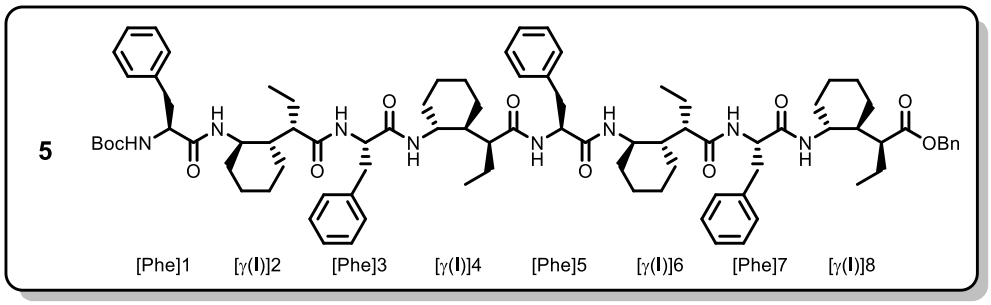


4

Atom 1	Atom 2	Distance	Designation
Reference distance			
Ph1	Ph2	2.80	
Sequential NOEs			
[Val]1(Hα)	[γ(I)]2(HN)	2.5	Strong
[Val]1(Hγ)	[γ(I)]2(HN)	3.9	Weak
[γ(I)]2(Hα)	[Val]3(HN)	2.5	Strong
[γ(I)]2(Et2)	[Val]3(HN)	4.1	Weak
[Val]3(Hα)	[γ(I)]4(HN)	2.5	Strong
[Val]3(Hγ)	[γ(I)]4(HN)	3.7	Weak
[γ(I)]4(Hα)	[Val]5(HN)	2.5	Strong
[Val]5(Hα)	[γ(I)]6(HN)	2.4	Strong
[γ(I)]6(Hα)	[Val]7(HN)	2.5	Strong
[γ(I)]6(Et2)	[Val]7(HN)	3.8	Weak
[γ(I)]8(Hα)	[Val]9(HN)	2.5	Strong
[γ(I)]8(Et2)	[Val]9(HN)	4.3	Weak
Non-sequential NOEs			
[Val]1(Boc)	[Val]3(HN)	4.8	Weak
[Val]1(Hα)	[Val]3(Hβ)	2.8	Strong
[Val]1(Hα)	[Val]3(HN)	3.2	Medium
[γ(I)]2(HN)	[Val]3(HN)	3.2	Medium
[Val]3(Hα)	[Val]5(HN)	3.3	Medium
[Val]3(Hα)	[Val]5(Hβ)	2.8	Strong
[γ(I)]4(HN)	[Val]5(HN)	3.6	Weak
[Val]5(Hα)	[Val]7(Hβ)	2.9	Strong
[Val]5(Hα)	[Val]7(HN)	2.7	Strong
[γ(I)]6(HN)	[Val]7(HN)	3.3	Medium
[Val]7(Hα)	[Val]9(HN)	3.6	Weak
[Val]7(Hα)	[Val]9(Hβ)	≈ 3.2[a]	Medium
[γ(I)]8(HN)	[Val]9(HN)	≈ 4.8[a]	Weak

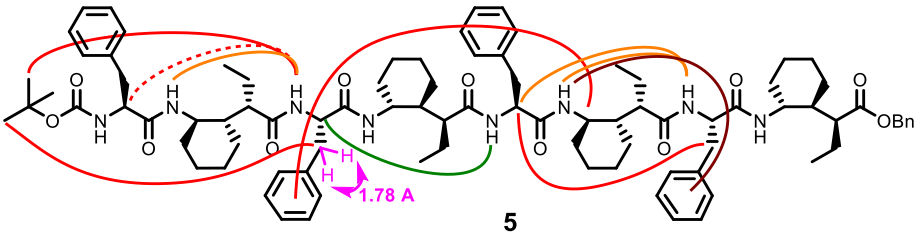
[a]Approximate value due to crosspeak overlap in ROESY spectrum.

Table S8. Resonance assignment of **5**



	Backbone				Sidechain	
Residue Number	**NH**	**γ**	**β**	**α**	**β2**	**γ3**
N-Terminus					Boc: 1.397	
[Phe]1	5.144			4.423	2.937, 3.035	
[γ(I)]2	6.868	3.581		2.156		0.703
[Phe]3	7.741			4.436	2.942, 3.070	
[γ(I)]4	6.537	3.475		2.119		0.701
[Phe]5	8.453			4.200	2.910, 3.022	
[γ(I)]6	6.233	3.501		2.188		0.663
[Phe]7	8.180			4.489	2.952, 3.137	
[γ(I)]8	5.859	3.636		2.449		0.789
C-Terminus					Bn: 5.010, 5.060	

Table S9. ROESY crosspeaks of **5**

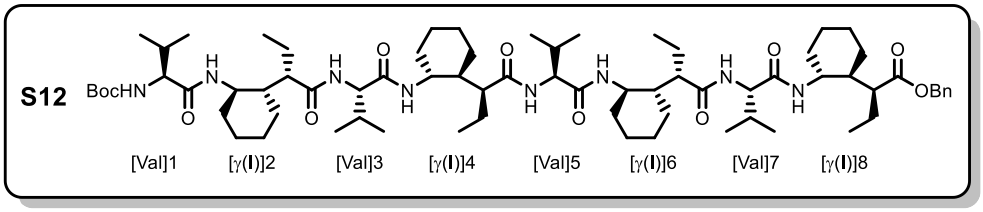


Atom 1	Atom 2	Distance	Designation
Reference distance			
[Phe]3(Hβ1)	[Phe]3(Hβ2)	1.78	
Sequential NOEs			
[Phe]1(Hα)	[γ(I)]2(HN)	2.3	Strong
[γ(I)]2(Hα)	[Phe]3(HN)	2.0	Strong
[γ(I)]2(Et2)	[Phe]3(Hα)	3.8	Weak
[γ(I)]2(Et2)	[Phe]3(HN)	4.0	Weak
[Phe]3(Hα)	[γ(I)]4(HN)	2.4	Strong
[γ(I)]4(Hα)	[Phe]5(Hβ1)	3.1	Medium
[γ(I)]4(Hα)	[Phe]5(HN)	2.0	Strong
[γ(I)]4(Et2)	[Phe]5(Hα)	4.0	Weak
[γ(I)]4(Et2)	[Phe]5(Hβ1)	3.7	Weak
[γ(I)]4(Et2)	[Phe]5(Hβ2)	3.9	Weak

[γ(I)]4(Et2)	[Phe]5(HN)	3.7	Weak
[Phe]5(Hα)	[γ(I)]6(HN)	2.1	Strong
[Phe]5(Hα)	[γ(I)]8(Bn)	3.4	Medium
[γ(I)]6(Hα)	[Phe]7(HN)	1.8	Strong
[γ(I)]6(Et2)	[Phe]7(HN)	4.5	Weak
[Phe]7(Hα)	[γ(I)]8(HN)	2.3	Strong
[Phe]7(HN)	[γ(I)]8(HN)	2.3	Strong
Non-sequential NOEs			
[Phe]1(Hα)	[Phe]3(HN)	≈ 5.0[a]	Weak
[Phe]1(Boc)	[Phe]3(Hβ1)	3.7	Weak
[Phe]1(Boc)	[Phe]3(HN)	4.1	Weak
[γ(I)]2(HN)	[Phe]3(HN)	3.4	Medium
[Phe]3(Hα)	[Phe]5(HN)	2.8	Strong
[Phe]3(Ph)	[γ(I)]6(Hγ)	3.5	Medium
[Phe]5(Hα)	[Phe]7(HN)	3.2	Medium
[Phe]5(Hα)	[Phe]7(Hβ2)	3.6	Weak
[γ(I)]6(HN)	[Phe]7(HN)	3.8	Weak
[γ(I)]6(HN)	[Phe]7(Ph)	6.1	Very weak

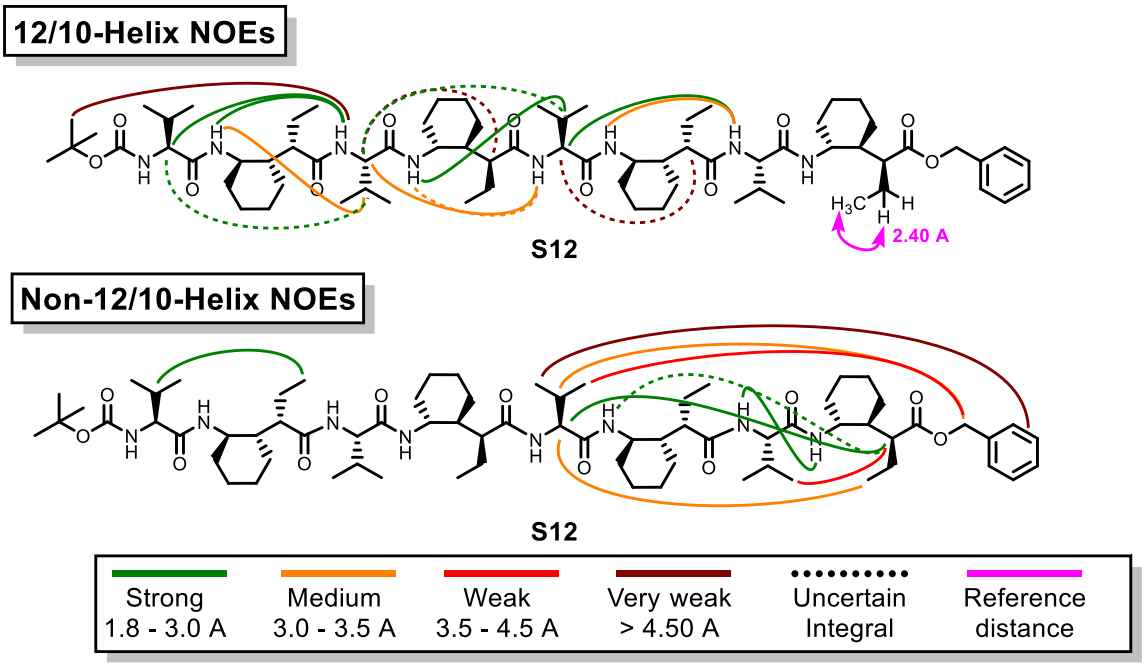
[a]Approximate value due to crosspeak overlap in ROESY spectrum.

Table S10. Resonance assignment of **S12**



	Backbone				Sidechain	
Residue Number	**NH**	**γ**	**β**	**α**	**β2**	**γ3**
N-Terminus					Boc: 1.442	
Val1	4.943			3.853	1.894	0.996
Gam2	7.079	3.644		2.140		0.759
Val3	7.787			4.061	1.919	1.015
Gam4	6.781	3.632		2.156		0.746
Val5	8.700			3.567	1.885	0.960
Gam6	6.741	3.729		2.489	1.495; 1.669	0.882
Val7	7.831			4.352	2.361	0.988
Gam8	6.090	3.647		2.553	1.550; 1.643	0.818
C-Terminus					Bn: 5.153	Ph: 7.329

Table S11. ROESY crosspeaks of **S12**



Atom 1	Atom 2	Distance	Designation
Reference Distance			
[γ(I)]6(Et2,1)	[γ(I)]6(Et2)	2.40	
Sequential NOEs			
[Val]1(Hα)	[γ(I)]2(HN)	2.1	Strong
[Val]1(Hγ)	[γ(I)]2(HN)	3.9	Medium
[γ(I)]2(Hα)	[Val]3(HN)	2.3	Strong
[γ(I)]2(Hα)	[Val]3(Hα)	3.0	Medium
[γ(I)]2(Et2)	[Val]3(HN)	3.6	Weak
[γ(I)]2(Et2)	[Val]3(Hα)	4.3	Weak
[Val]3(Hα)	[γ(I)]4(HN)	2.2	Strong
[Val]3(Hγ)	[γ(I)]4(HN)	2.8	Strong
[γ(I)]4(Hα)	[Val]5(HN)	2.4	Strong
[γ(I)]4(Hα)	[Val]5(Hα)	3.5	Weak
[γ(I)]4(Et2)	[Val]5(HN)	4.5	Weak
[γ(I)]4(Et2)	[Val]5(Hα)	3.0	Medium
[Val]5(Hα)	[γ(I)]6(HN)	2.2	Strong
[Val]5(Hγ)	[γ(I)]6(HN)	3.8	Weak
[γ(I)]6(Hα)	[Val]7(HN)	2.1	Strong
[γ(I)]6(Et2)	[Val]7(HN)	3.8	Weak
[Val]7(Hα)	[γ(I)]8(HN)	2.7	Strong
[Val]7(Hβ)	[γ(I)]8(HN)	3.1	Medium
Non-sequential 12/10-helical NOEs			
[Val]1(Boc)	[Val]3(HN)	4.5	Weak
[Val]1(Hα)	[Val]3(HN)	2.8	Strong
[Val]1(Hα)	[Val]3(Hβ)	2.2	Strong
[γ(I)]2(HN)	[Val]3(HN)	2.9	Strong
[γ(I)]2(HN)	[Val]3(Hβ)	3.0	Medium
[Val]3(Hα)	[γ(I)]4(Hα)	3.1	Medium
[Val]3(Hα)	[Val]5(HN)	3.0	Medium
[Val]3(Hα)	[Val]5(Hβ)	2.4	Strong

[γ(I)]4(HN)	[Val]5(HN)	3.2	Medium
[γ(I)]4(HN)	[Val]5(Hβ)	2.8	Strong
[Val]5(Hα)	[γ(I)]6(Hα)	3.3	Medium
[Val]5(Hα)	[Val]7(HN)	3.4	Medium
[γ(I)]6(HN)	[Val]7(HN)	3.0	Medium
Non-sequential, non-12/10-helical NOEs			
[Val]5(Hα)	[γ(I)]8(Hα)	2.9	Strong
[Val]5(Hα)	[γ(I)]8(Et2)	3.4	Medium
[Val]5(Hα)	[γ(I)]8(Bn)	3.3	Medium
[Val]5(Hγ)	[γ(I)]8(Bn)	4.0	Weak
[Val]5(Hγ)	[γ(I)]8(Ph)	4.9	Weak
[γ(I)]6(HN)	[γ(I)]8(Hα)	2.4	Strong
[γ(I)]6(HN)	[γ(I)]8(Bn)	3.8	Weak
[Val]7(HN)	[γ(I)]8(HN)	2.8	Strong
[Val]7(Hγ)	[γ(I)]8(Hα)	3.5	Weak

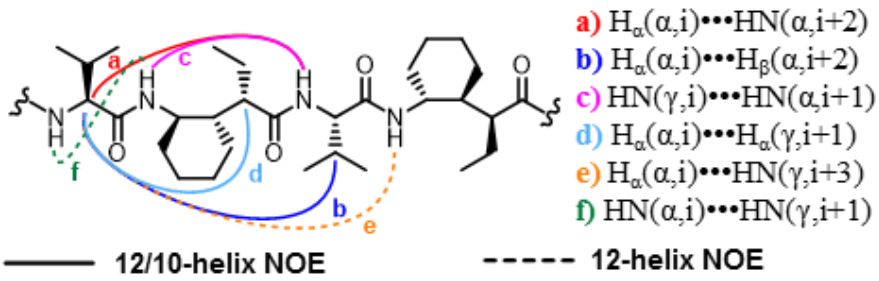
e. NOE Pattern Analysis

12/10-Helical NOEs vs. Non-12/10-Helical NOEs of Foldamers Containing C-terminal γ(I)

Most non-sequential NOEs of compounds **2a**, **S11**, **4**, **5**, and **S12** were consistent with the 12/10-helical structure as determined by comparison with crystal **2a** and model helices (**Table S12**). NOEs involving C-terminal residues [γ(**I**)]6 in compound **S11** and residue [γ(**I**)]8 in compound **S12** are not expected for an ideal 12/10-helix adopted uniformly along the foldamer sequence, however, raising a few possibilities: that these peptides prefer a different type of helix structure; that they exhibit significant conformational heterogeneity; or that the C-terminal residues adopt a backbone conformation distinct from those of the other residues comprising the 12/10-helical foldamer. Although crystallographic evidence points to the 12/10-helix as the preferred secondary structure of α/γ(**I**)-peptides, it is not *a priori* certain that this trend follows to the solution phase or to longer foldamers. Short α-peptides are known to have a "split personality" in their population of both the 3_{10}- and α-helix in solution,[S12] and length-dependent conformational switching has been reported in hybrid α/β-peptides forming mixed[S13] and polar[S14] helices. Careful consideration of the 2D-NMR data in relation to model helices (*vide supra*) would be useful in evaluating the possibilities.

Table S12 below shows a comparison of the NOE-calculated interproton distances between protons in non-sequential residues of **S12** with the crystal of **2a** and interproton distances found in the model helices. The NOE pattern clearly indicates that the 20/18 helix is not the preferred solution secondary structure of **S12**. The crosspeaks observed correlate poorly with those expected for a 20/18-helical structure, and the expected crosspeaks are not detected. Interproton distances **a**, **b**, and **d** are similar in the model 12- and 12/10-helices, thus these experimental distances cannot distinguish the two structures unambiguously. Distance **c** points more to the 12/10-helix than the 12-helix. Strong and medium crosspeaks of **S12** are detected, although a weak crosspeak would have been expected for the 12-helix. The absence of distance **f** in the ROESY data is particularly strong evidence against the 12-helix. Distance **f** would be a sensitive probe of 12-helical secondary structure given the short spatial and atom connectivity distance between the protons, which are separated by only ≈2.7 Å and four atoms. The ROESY data indicate that the 12-helix is not the preferred secondary structure of **S12** and also suggest that the 12-helix is not highly populated.

Table S12. Comparison of crystal **2a**, model **S16** helices, and **S12** NOE-derived interproton distances



	2a Crystal 12/10-Helix	Model S16 12/10-helix	Model S16 12-helix	Model S16 20/18-helix	S12 NOE distances
Distance a	2.78	2.66	3.09	4.70	3.03[a]
Distance b	2.47	2.49	3.16	6.89	2.40[b]
Distance c	2.69	2.82	4.02	4.38	2.87[c]
Distance d	4.14	3.92	4.07	4.65	4.0-4.5[d]
Distance e	5.02	5.22	3.08	5.12	ND
Distance f	4.39	4.66	2.73	4.19	ND

[a][Val]3(Hα)−[Val]5(Hα)
[b][Val]3(Hα)−[Val]5(Hβ)
[c][γ(**I**)]2(HN)−[Val]3(HN)
[d][Val]5(Hα)−[γ(**I**)]6(Hα). Approximate distance due to crosspeak overlap.

The non-12/10-helical NOEs—those involving the C-terminal γ(**I**)-residue and not expected for an idealized 12/10-helix—point most strongly to a specific alternative conformation of this residue. This would not be unexpected, given the tendency of short peptides to fray at the termini and the fact that residue [γ(**I**)]8 cannot donate or accept 12/10-helical H-bonds. Still, the fact that the shortest non-sequential NOE involves this non-12/10-helical residue is curious, and NMR-restrained simulated annealing calculations were used to characterize the conformational properties of this residue in **S12**.

f. NMR-Restrained Simulated Annealing Calculations of 2a, S11, S12, 4, and 5

NMR Calculation Parameters

The internuclear distances above, including sequential and non-sequential distances (but not intraresidue), were used as distance restraints in simulated annealing calculations of **2a**, **S11**, **S12**, **4**, and **5** using the Crystallography and NMR System (CNS v1.3) software package.[S15] Molecular topologies and parameters for the Boc and benzyl ester protecting groups were generated using the PRODRG2 Server.[S16] Topology for the BrBn ester protecting group was built manually, and parameters were generated using the ACPYPE script[S17] operating the ANTECHAMBER package of AmberTools14.[S18] Parameters for the γ(**I**)-residue were built manually, as were the linkage definitions needed to attach the protecting groups, γ-residues, and α-residues together to form the foldamers.

Dihedral restraints were added to enforce chair conformations of the γ(**I**)-residue cyclohexyl ring. Boat and twist-boat conformations were frequently found among the trial structures in initial calculations (**Figure S5a**). The tendency of CNS to find thermodynamically unstable cyclohexyl conformations has been reported by our group[S19] and others.[S20] We found that adding one or two generous dihedral angle restraints to each cyclohexyl ring successfully directed the cyclohexyl rings toward the expected chair conformation (**Figure S5b**). We do not believe that these restraints could significantly bias the trajectory of the simulations. A back-of-the-envelope calculation summing the energetic penalties of twist-boat adoption and approximate A-values of the *trans*-diaxial cyclohexane substituents has the twist-boat of γ(**I**) about 7-8 kcal mol^{-1} higher in energy than the chair conformation. All final accepted structures featured no energetic penalty due to the dihedral restraints, thus the restraint did not influence the choice of accepted structures.

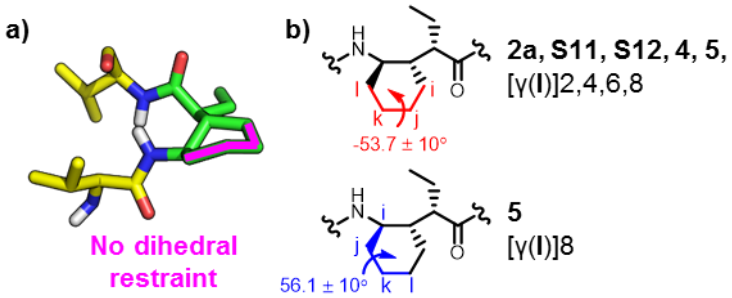


Figure S5. a) Twist-boat conformation produced in simulated annealing calculations of **S12** when no dihedral restraints are employed. **b)** Dihedral restraints used in simulated annealing calculations. Residue [γ(**I**)]8 in foldamer **5** required two dihedral restraints to prevent twist-boat formation.

Molecular topology files (.mtf) were generated from the primary sequence using the generate-seq.inp input file included in the CNS software package. The molecular topology files were used as the input to construct extended structures (.pdb) using the generate-ext.inp input file. The .mtf file, the extended .pdb file, the distance restraint table, and a dihedral restraint table were the input data in the simulated annealing calculations using anneal.inp as the input file for 50 or 1000 trial structures. The hot stage was run at 10,000 K for 15 ps in 1000 torsion steps, using 0.1 Van der Waals scale factor, 150 NOE scale factor, and 100 dihedral restraint scale factor. The 1st cooling stage was run from 10,000 K to 0 K in 1000 torsion steps and a 50 K temperature step, using 0.1 →1.0 Van der Waals scale factor, 150 NOE scale factor, and 200 dihedral restraint scale factor. The 2nd cooling stage was run from 300 K to 0 K for 15 ps in 3000 Cartesian steps with a 25 K temperature step, using 1.0→4.0 Van der Waals scale factor, 150 NOE scale factor, and 200 dihedral restraint scale factor. A final minimization stage with 10 cycles of 200-step minimization with a 75 NOE scale factor and 400 dihedral scale factor. The 10 structures of the 500 or 1000 trial structures with the lowest overall energy were selected with the accept.inp input file.

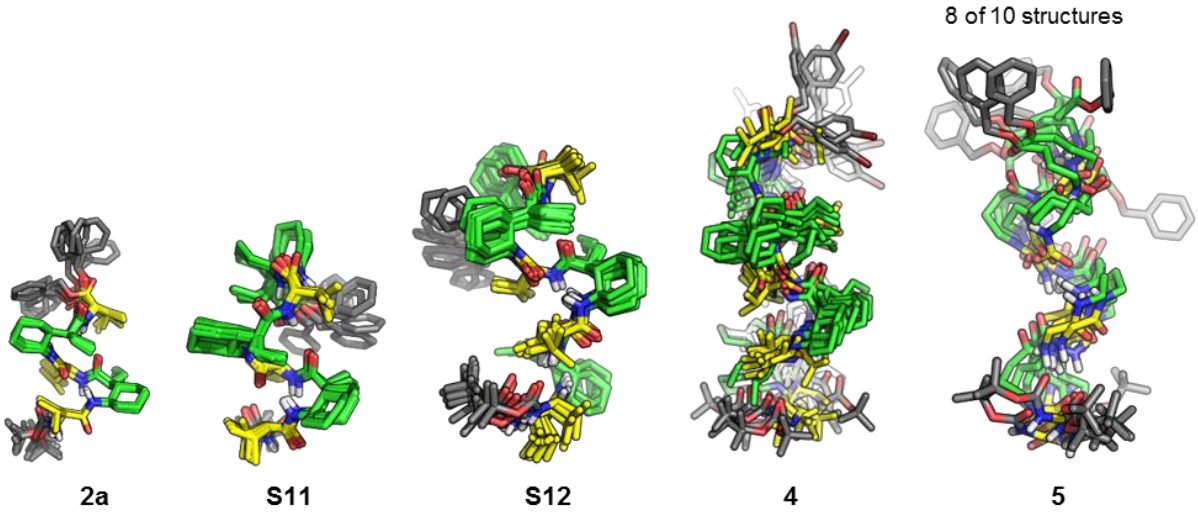
Accepted structures were visualized using PyMOL v1.7.0.3.[S11] Alignment of ensemble structures to crystal structures was performed using the "align" command of PyMOL.

Table S13. CNS simulated annealing calculation statistics for **2a**, **S11**, **S12**, **4**, and **5**

	2a	**S11**	**S12**	**4**	**5**
Trial structures	1000	1000	500	1000	1000
Distance restraints					

Total NOE	14	27	35	24	20
Intra-residue	0	1	0	0	0
Inter-residue	14	26	35	24	20
Sequential	8	13	17	12	16
Non-sequential	6	13	18	12	8
Hydrogen bond restraints	0	0	0	0	0
Dihedral restraints	2	3	4	5	5
Restraint violations					
Distance restraint violations > 0.5 Å	0.00 ± 0.00	0.80 ± 0.42	0.20 ± 0.422	0.00 ± 0.00	0.00 ± 0.00
Dihedral restraint violations > 5°	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Deviations from idealized geometry					
Bond lengths (Å)	0.010 ± 0.000	0.011 ± 0.000	0.012 ± 0.000	0.011 ± 0.000	0.010 ± 0.000
Bond angles (°)	0.820 ± 0.001	0.924 ± 0.015	0.967 ± 0.019	0.859 ± 0.014	0.761 ± 0.023
Impropers (°)	0.550 ± 0..005	0.576 ± 0.106	0.561 ± 0.011	0.483 ± 0.184	0.381 ± 0.059
Average pairwise RMSD of 10 lowest-energy structures (Å)					
Backbone	0.130 ± 0.073	0.371 ± 0.259	0.775 ± 0.284	2.00 ± 0.76	2.01 ± 0.59
Heavy atom	1.90 ± 0.63	1.32 ± 0.49	1.78 ± 0.55	3.14 ± 0.92	4.17 ± 1.01

NMR Simulated Annealing Calculation Structures for 2a, S11, S12, 4, and 5



Compound		1	2	3	4	5	6	7	8	9	
2a	Boc -	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]					- OBn
S11	Boc -	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]				- OBn
S12	Boc -	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]		- OBn
4	Boc -	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	[γ(**I**)]	[Val]	- OBrBn
5	Boc -	[Phe]	[γ(**I**)]	[Phe]	[γ(**I**)]	[Phe]	[γ(**I**)]	[Phe]	[γ(**I**)]		- OBn

Figure S6. Accepted 10 lowest-energy structures from simulated annealing calculations for peptides **2a**, **S11**, **S12**, and **4**, with 8 of 10 lowest-energy structures with sidechains removed for clarity of **5**. α-Residues are depicted in yellow, γ-residues in green, and protecting groups in gray.

The 10-lowest-energy NMR structures of hexapeptide **S11** and octapeptide **S12**, the two foldamers with C-terminal γ(**I**)-residues, depict relatively tightly clustered backbone RMSDs which reveal a few unusual characteristics of the structures (**Figure S7a**). Seen most clearly in longer octapeptide **S12**, the helix curvature is greater than that seen in 12/10-helix crystal structures (**Figure S7b**). As shown at in **Table S11**, the two C-terminal residues of both foldamers feature non-12/10-helical NOE patterns, and in the NMR ensembles these residues adopt conformations unlike the residues participating in the 12/10-helix (**Figure S7c**). The residue [Val]7 in **S12** adopts α-helix-like ϕ/ψ values enforced by the large ROESY crosspeak between HN(α,i) and HN(γ,i+1) from [Val]7 to [γ(**I**)]8 (distance **f** in **Table S12**). Another feature of the C-terminus in the calculated structures is a C15 H-bond from the benzyl ester C=O to the H-N of [γ(**I**)]6. Since this H-bond acceptor is relatively weak, it is possible that this is an artifact of the calculations.

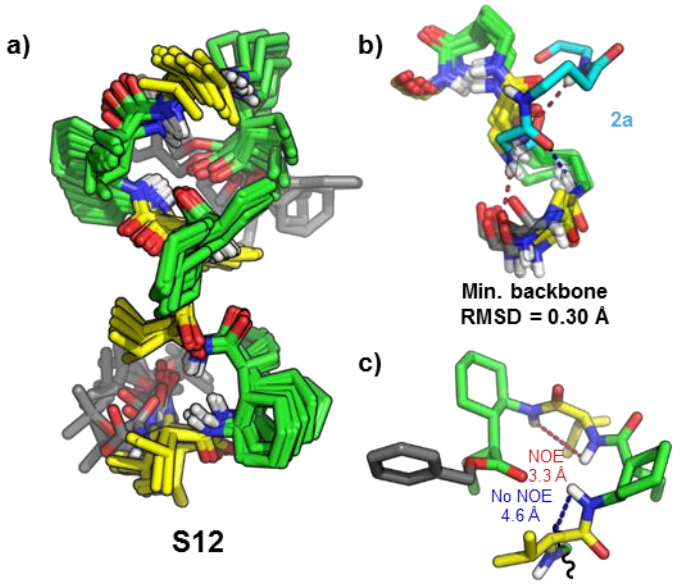
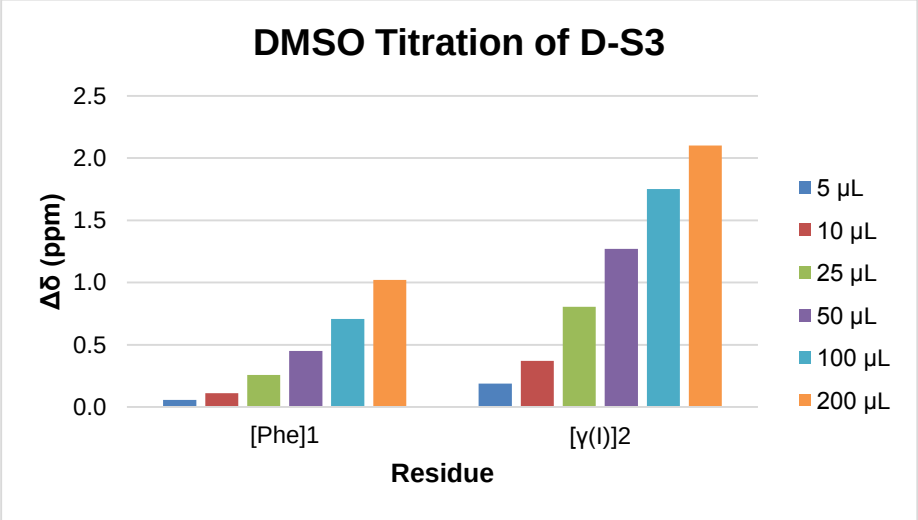
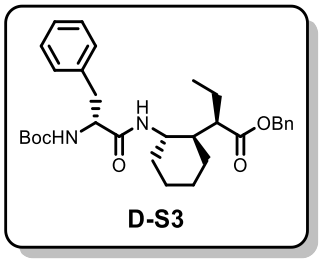


Figure S7. a) 10 lowest-energy NMR structures of **S12**. Protecting groups are shown in gray, α-residues in yellow, and γ-residues in green. **b)** Backbone overlay of the first three residues of the **S12** NMR ensemble and crystal structure **2a**. **c)** C-terminus of minimized average structure, depicting the non-12/10-helical distance **f** (see **Table S12**) for 12/10-helical [Val]5 and [γ(**I**)]6 (no NOE) and for non-12/10-helical [Val]7 and [γ(**I**)]8 (NOE detected).

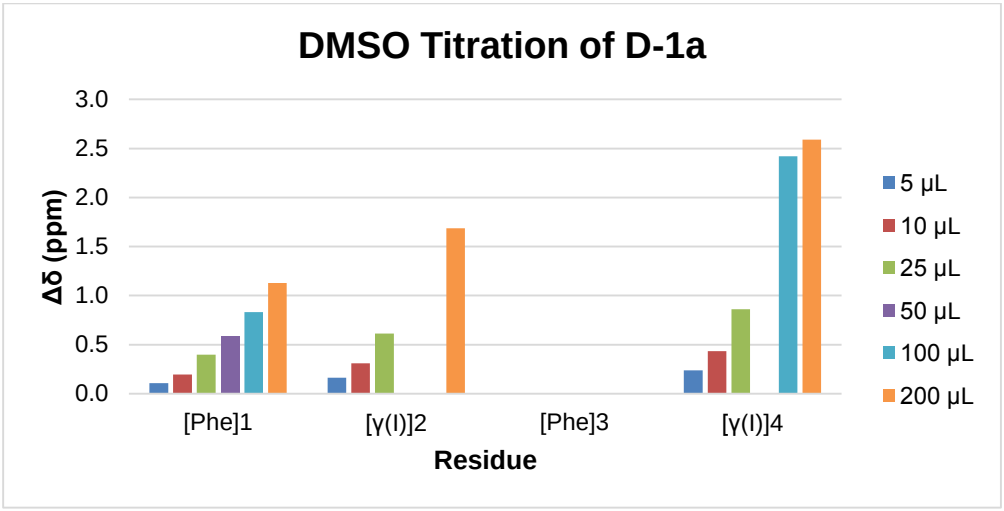
V. DMSO Titrations

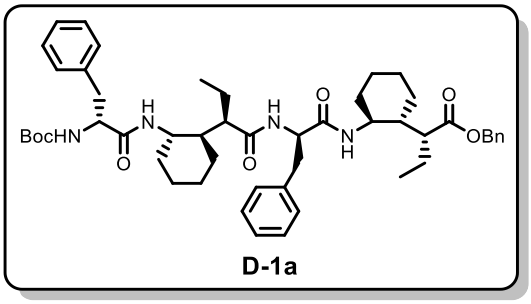
Peptides were dissolved to 1 mM in 500 µL dry $CDCl_3$ and transferred to a 5 mm NMR tube sealed with a rubber septum and Teflon tape. DMSO-d_6 was injected via a Hamilton syringe, the NMR tube was inverted 5 times and shaken to ensure homogeneity, and the ^{1}H-NMR acquired on a Bruker Avance III 500 MHz spectrometer with 32 scans.



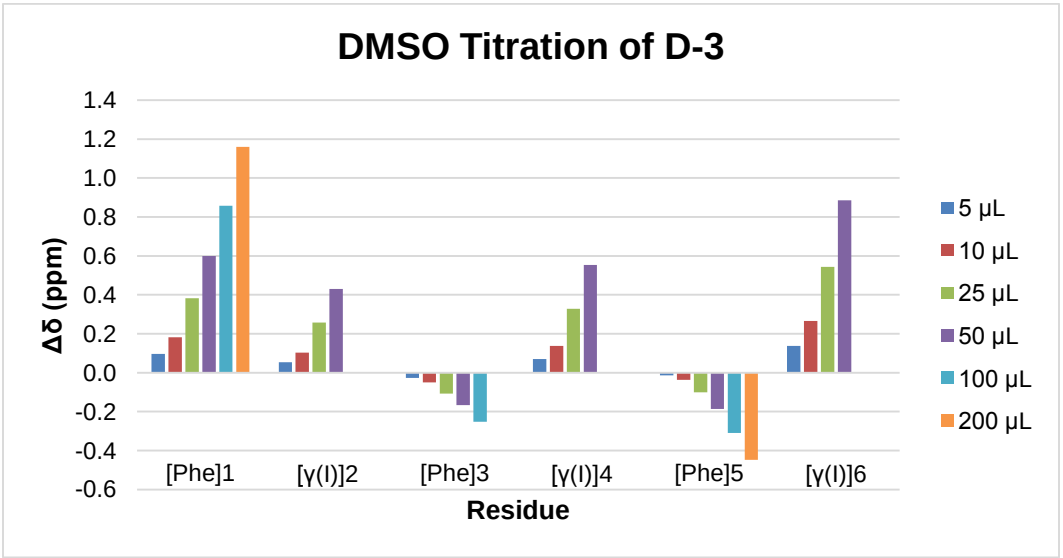

BocHN
H
N
O
O
OBn
O

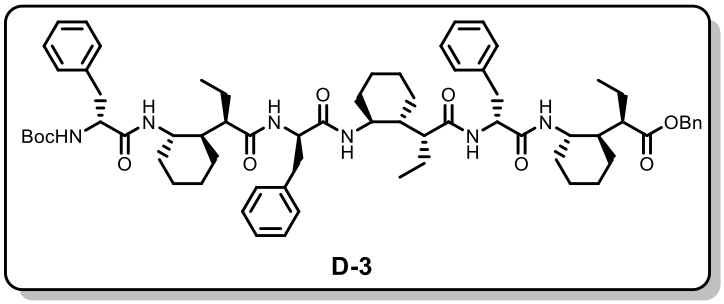
D-S3


DMSO Titration of D-1a
Δδ (ppm)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
[Phe]1
[γ(I)]2
[Phe]3
[γ(I)]4
Residue
5 µL
10 µL
25 µL
50 µL
100 µL
200 µL

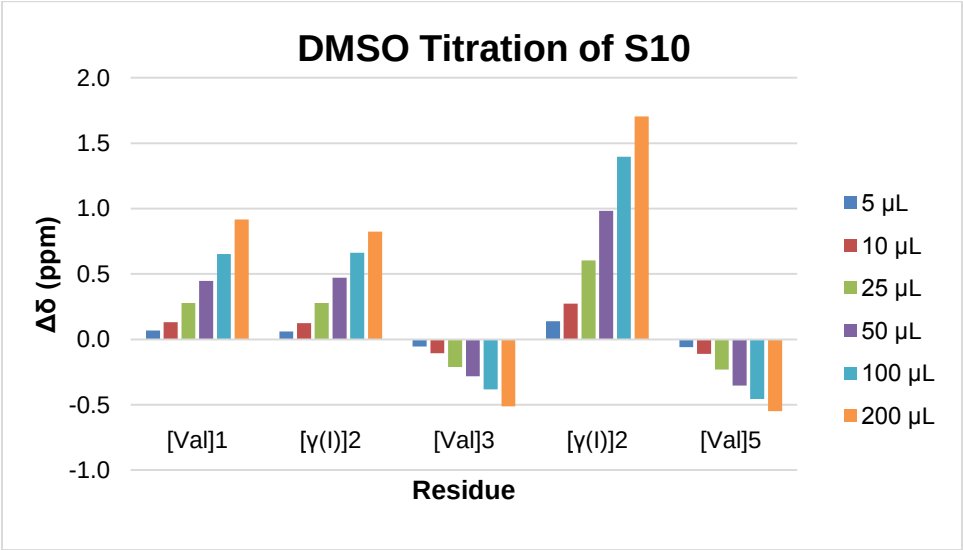

BocHN
H
N
O
H
N
O
O
N
H
O
OBn

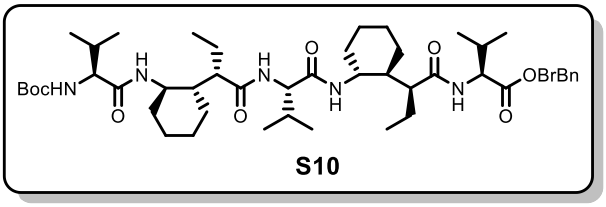
D-1a


DMSO Titration of D-3
Δδ (ppm)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
[Phe]1
[γ(I)]2
[Phe]3
[γ(I)]4
[Phe]5
[γ(I)]6
Residue
5 µL
10 µL
25 µL
50 µL
100 µL
200 µL

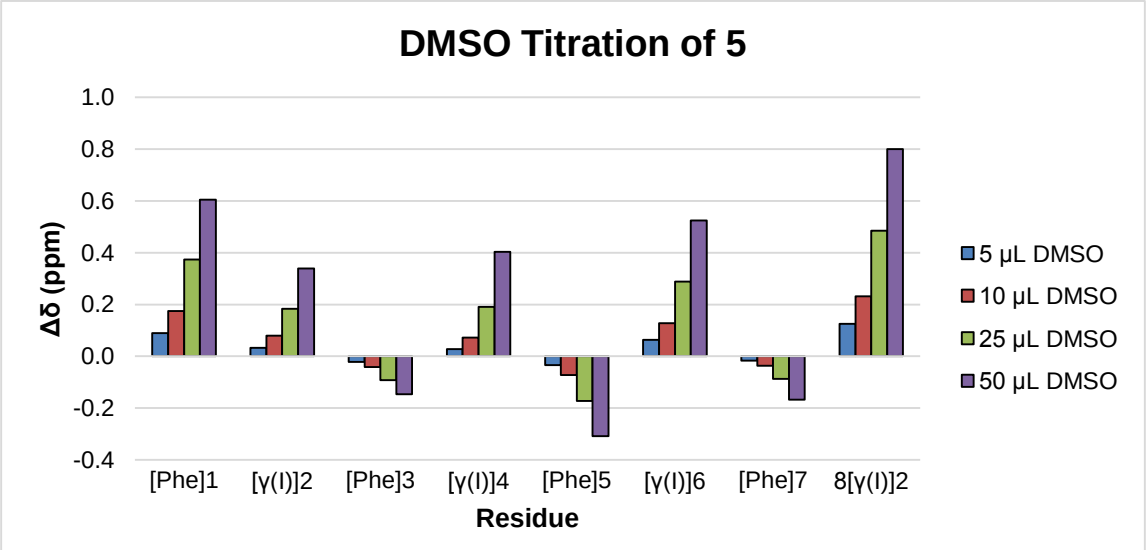

BocHN
OBn

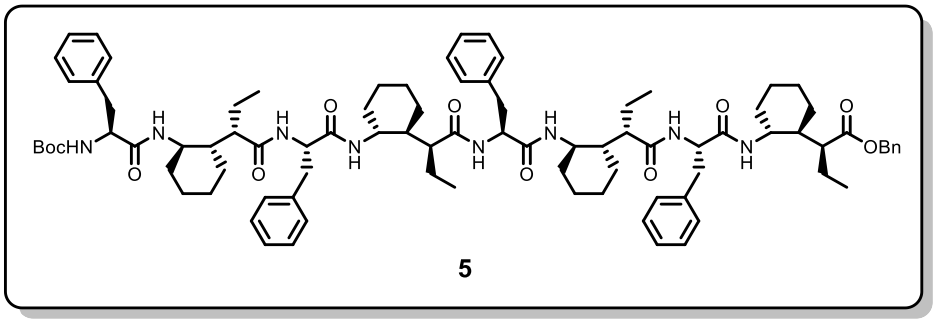
D-3


DMSO Titration of S10
2.0
1.5
1.0
0.5
0.0
-0.5
-1.0
Δδ (ppm)
[Val]1
[γ(l)]2
[Val]3
[γ(l)]2
[Val]5
Residue
5 μL
10 μL
25 μL
50 μL
100 μL
200 μL


BocHN
OBrBn

S10


DMSO Titration of 5
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
Δδ (ppm)
[Phe]1
[γ(l)]2
[Phe]3
[γ(l)]4
[Phe]5
[γ(l)]6
[Phe]7
8[γ(l)]2
Residue
5 μL DMSO
10 μL DMSO
25 μL DMSO
50 μL DMSO



5

VI. Hydrogen-Deuterium Exchange Experiments

Peptides were dissolved to 1 mM in 500 μL $CDCl_3$ purified by passage through basic alumina and stored over 3 Å molecular sieves, then transferred to a 5 mm NMR tube sealed with a rubber septum and Teflon tape. CD_3OD was injected via a Hamilton syringe, the NMR tube was inverted 5 times and shaken to ensure homogeneity, and ^{1}H-NMR data were collected at regular intervals at 24°C on a Bruker Avance III 500 MHz spectrometer using the kin1d pulse program, d1 = 1 sec, and ns = 32 scans. Data were batch processed with MestReNova. Spectra were automatically phased, visually inspected, and manually phased as needed. Baseline correction was done using the ablative baseline correction algorithm. Integrals were defined manually. Half-lives were calculated by fitting an exponential decay function to the data using Microsoft Excel. Integrals below 0.09 were too near the baseline for reliable integration and excluded in the fit.

Table S14. HDX Integral Data[a] for **5**

Elapsed time (hr)	[Phe]1	[γ(I)]2	[Phe]3	[γ(I)]4	[Phe]5	[γ(I)]6	[Phe]7	[γ(I)]8
0.00								
0.06	0.765	0.674	0.794	0.652	0.709		0.633	0.590
0.09	0.750	0.671	0.784	0.624	0.687		0.621	0.581
0.12	0.746	0.674	0.784	0.635	0.701		0.636	0.588
0.21	0.702	0.666	0.770	0.637	0.690		0.613	0.565
0.29	0.679	0.674	0.775	0.632	0.697		0.626	0.569
0.37	0.646	0.659	0.776	0.628	0.682		0.625	0.563
0.46	0.616	0.657	0.764	0.625	0.689		0.619	0.570
0.54	0.603	0.662	0.760	0.620	0.682		0.621	0.570
0.62	0.587	0.669	0.775	0.630	0.696		0.616	0.572
0.71	0.565	0.652	0.768	0.617	0.687		0.619	0.573
0.79	0.532	0.646	0.743	0.610	0.679		0.603	0.565
0.87	0.515	0.659	0.765	0.626	0.682		0.609	0.579
0.96	0.511	0.663	0.758	0.620	0.681		0.611	0.563
1.06	0.483	0.640	0.737	0.605	0.680		0.605	0.563
1.16	0.471	0.650	0.755	0.614	0.684		0.598	0.566
1.32	0.442	0.644	0.753	0.613	0.678		0.603	0.565
1.49	0.414	0.645	0.746	0.614	0.679		0.600	0.562
1.66	0.382	0.648	0.751	0.616	0.665		0.597	0.541
1.82	0.362	0.645	0.754	0.614	0.681		0.594	0.554
1.99	0.338	0.629	0.735	0.595	0.671		0.588	0.559
2.16	0.320	0.640	0.742	0.602	0.676		0.588	0.548
2.41	0.302	0.650	0.738	0.605	0.676		0.598	0.553
2.66	0.268	0.643	0.736	0.612	0.679		0.580	0.552
2.91	0.245	0.636	0.720	0.605	0.669		0.570	0.544
3.16	0.226	0.628	0.715	0.600	0.672		0.575	0.541

3.41	0.203	0.626	0.721	0.594	0.666	0.565	0.529
3.66	0.196	0.625	0.717	0.602	0.660	0.569	0.542
3.91	0.184	0.625	0.718	0.603	0.666	0.565	0.531
4.16	0.163	0.629	0.715	0.597	0.666	0.557	0.523
4.67	0.149	0.621	0.706	0.596	0.657	0.552	0.523
5.17	0.137	0.627	0.705	0.598	0.656	0.548	0.528
5.67	0.110	0.609	0.694	0.586	0.651	0.543	0.516
6.17	0.105	0.617	0.697	0.591	0.656	0.534	0.512
7.17	0.095	0.611	0.691	0.597	0.654	0.520	0.497
8.17	0.074	0.609	0.679	0.592	0.653	0.507	0.493
9.17	0.070	0.604	0.673	0.588	0.645	0.502	0.477
10.17	0.067	0.591	0.662	0.586	0.643	0.490	0.463
13.54		0.608	0.594	0.597	0.587	0.466	0.430
18.34		0.586	0.567	0.598	0.573	0.424	0.393
21.62		0.573	0.542	0.591	0.567	0.393	0.360
21.69		0.559	0.536	0.576	0.560	0.386	0.362
30.96		0.550	0.523	0.577	0.551	0.335	0.307
35.96		0.504	0.498	0.543	0.531	0.299	0.283
54.06		0.463	0.463	0.522	0.495	0.222	0.210
65.82		0.403	0.421	0.491	0.454	0.169	0.168
66.97		0.415	0.431	0.497	0.463	0.177	0.169
74.57		0.377	0.407	0.476	0.438	0.150	0.145
88.81		0.359	0.389	0.463	0.412	0.138	0.107
99.11		0.333	0.363	0.446	0.390	0.116	0.080
108.69		0.317	0.337	0.426	0.378	0.116	0.083
120.06		0.291	0.326	0.407	0.346	0.098	0.063
133.47		0.259	0.301	0.373	0.322	0.093	0.059
148.07		0.244	0.275	0.364	0.292	0.095	0.053

[a]Integration value relative to benzyl proton at δ 5.11 ppm = 1. [b]Chemical shift overlap with phenyl protons prevented reliable integration.

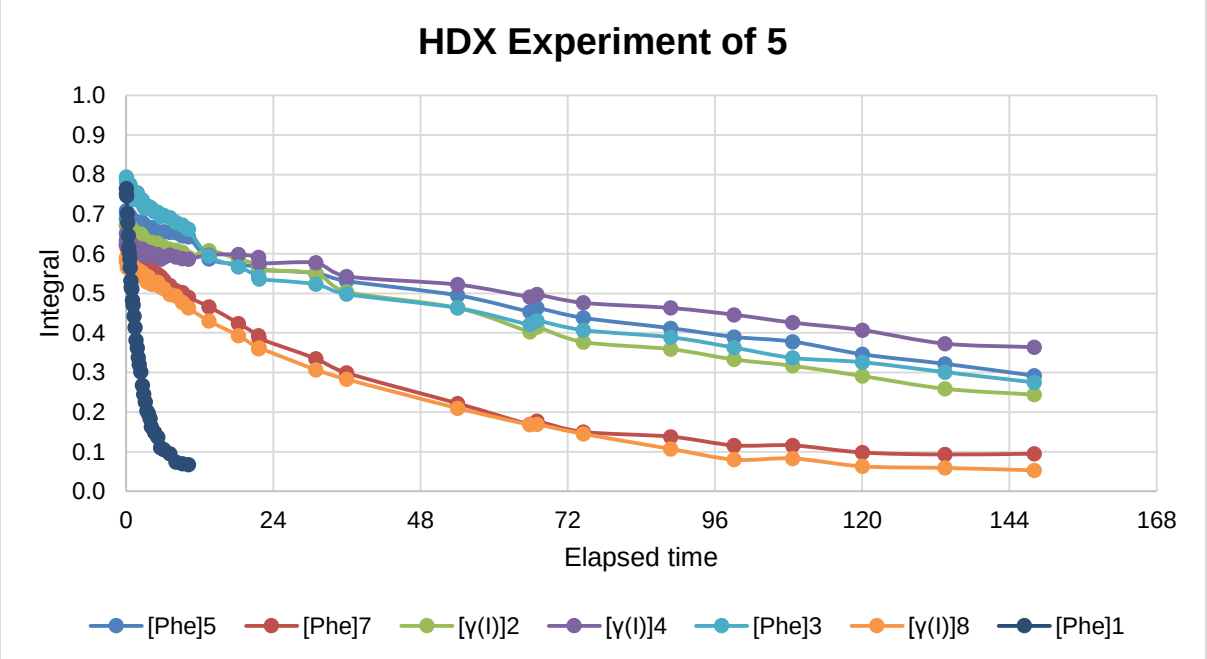


Figure S8. HDX data for peptide **5**.

Table S15. HDX integral data[a] for **6**

Elapsed time (hr)	[Phe]1	[γ(I)]2
0.06	0.890	0.943
0.16	0.836	0.938
0.26	0.832	0.938
0.34	0.780	0.873
0.42	0.730	0.903
0.51	0.651	0.867
0.59	0.673	0.913
0.67	0.606	0.885
0.76	0.547	0.879
0.84	0.531	0.818
0.92	0.476	0.830
1.01	0.467	0.817
1.39	0.376	0.759
1.64	0.336	0.788
1.89	0.312	0.803
2.14	0.256	0.749
2.39	0.243	0.742
2.64	0.207	0.701
2.89	0.195	0.719
3.14	0.125	0.648
3.39	0.155	0.667
3.64	0.123	0.629
3.89	0.143	0.633
4.14	0.102	0.632
4.69	0.084	0.564
5.69	0.039	0.492
6.69	0.000	0.454
7.69		0.400
8.69		0.425
9.69		0.400
10.69		0.322
11.69		0.358
12.69		0.304
13.69		0.268
21.41		0.152

[a]Integration value relative to valine H_α at δ 4.56 ppm = 1.

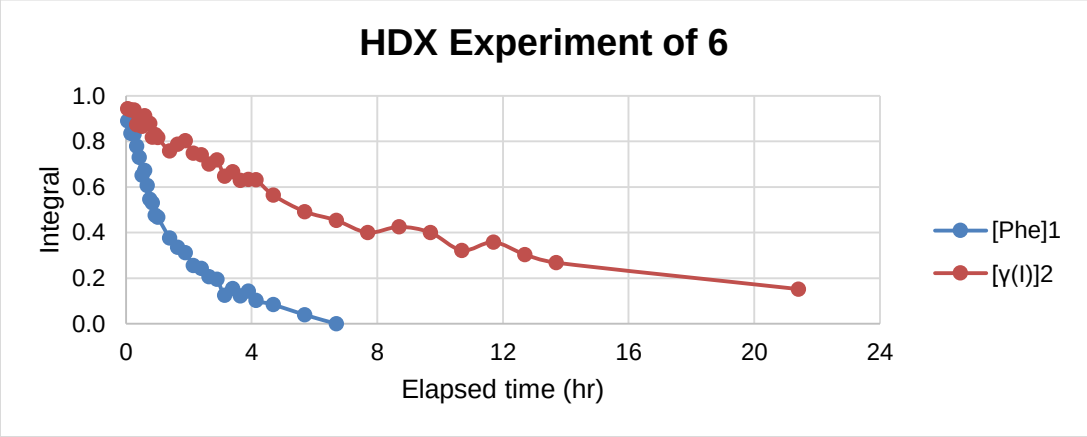


Figure S9. HDX data for peptide **6.**

Design of Model Peptides for HDX

HDX half-lives of backbone amide NHs within short peptides without tertiary structure are mostly influenced by three factors: 1) the intrinsic half-life of the amino acid residue's amide NH; 2) the peptide's primary structure; that is, the steric and electronic influence of residues adjacent to the backbone amide NH; and 3) the peptide's secondary structure; that is, the HDX protection offered by intramolecular H-bond engagement. The HDX experiments reported here seek to measure the influence of only the third factor on the relative HDX rate of the two amide types—NH_α forming 12-membered H-bonds, and NH_γ forming 10-membered H-bonds—within α/γ(**I**)-peptides. Since all three factors will differ for NH_α of Phe and NH_γ of γ(**I**), a model compound approximating the first two factors for each residue must be used. In other words, the model compound must include both Phe and γ(**I**) (factor 1 included), include the relevant proximal steric bulk around each amide (factor 2 included), and feature no strong H-bonding (factor 3 excluded). Any increase in HDX half-life from this model compound is attributed to an increase in H-bond engagement of the amide NH.

For amide NH_α of Phe, the residues on either side of the Phe are likely to have an impact on HDX half-life. Model compounds **S14** and **S15** were assessed to test the validity of exchanging the group N-terminal of NH_α from γ(**I**) to 2-ethylbutyryl in design of the final model compound (**Figure S10**). The 2-ethylbutyryl group is more desirable since it contains fewer H-bond donors/acceptors than the γ(**I**)-residue, and removing the possibility of intramolecular H-bonding will ensure that factor 3 is not operative in increasing HDX half-life of the model compound. The HDX half-lives of NH_α in **S14** and **S15** were found to be 0.52 hr and 0.59 hr respectively, indicating that 2-ethylbutyryl is a good mimic of γ(**I**) for HDX studies.

Model compound **6** incorporates the 2-ethylbutyryl group N-terminal of Phe and γ(**I**) C-terminal of it. One intramolecular H-bond could plausibly be formed (**Figure S10**), although this H-bond type, C=O(γ,i)→H-N(α,i−1) has not been observed crystallographically. It should be noted that, if this H-bond is formed, Factor 3 will influence the HDX half-life of NH_α in the model compound and thus will be longer. A relative increase from this HDX half-lfie to the HDX half-life of the full-length α/γ(**I**)-peptide will thus be underestimated. Since this H-bond type is expected to be the stronger of the two, the potential H-bond in **6** would not strengthen the conclusions from the HDX data.

Compound **6** also features NH_γ in an environment similar to its context in α/γ(**I**)-peptides. N-terminal of γ(**I**) is Phe, and the C-terminus is a benzyl ester instead of an amide in order to reduce the number of good H-bond acceptors and H-bond donors. Since this C-terminus is spatially distant from NH_γ, it should not have a large impact on HDX rate via factor 2.

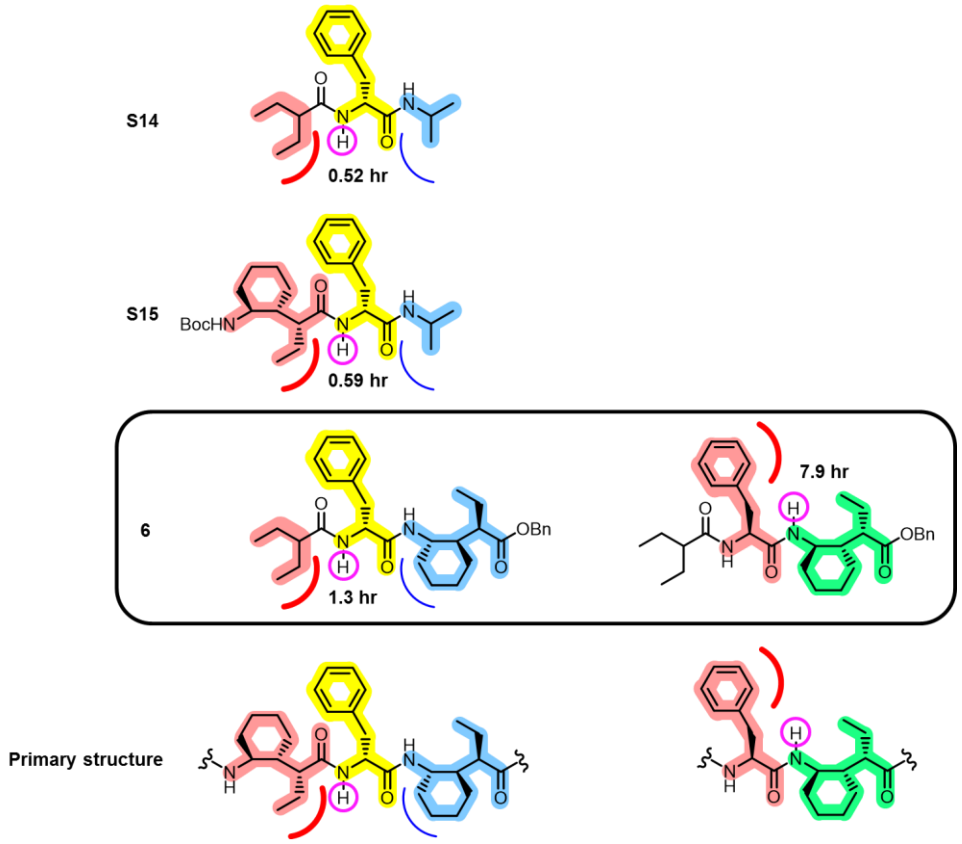
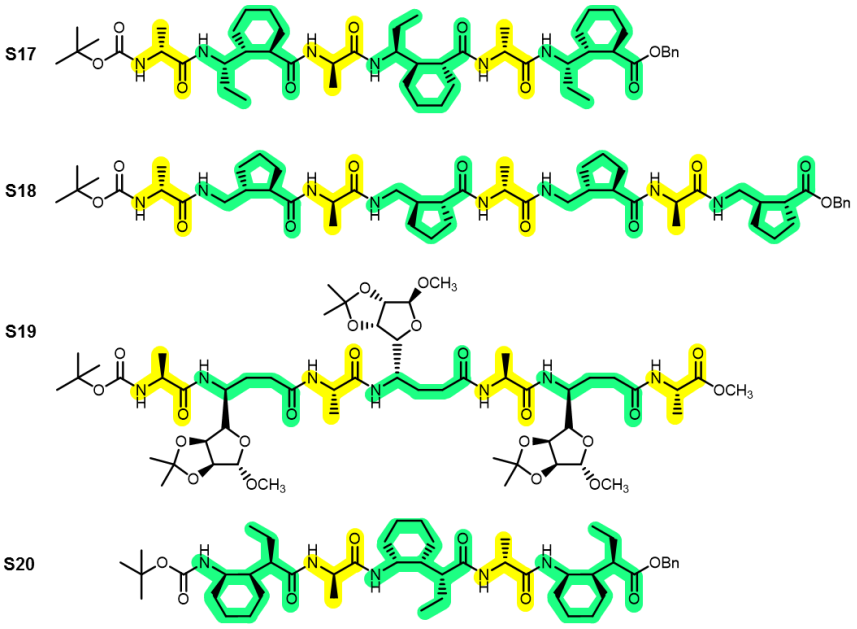


Figure S10. HDX model compounds. **a)** Model compounds **S14** and **S15**, used to assess 2-ethylbutyryl group as γ(**I**) mimic in terms of its ability to influence HDX rate of NH_{α}. HDX half-lives are shown next to NH_{α}. **b)** Model compound **6**, which was used as a reference for HDX experiments of full-length α/γ(**I**)-peptides, with potential H-bond shown in light blue. HDX half-lives are shown next to their respective NH. **c)** Depiction of α/γ(**I**)-peptide primary context of each residue relevant to influencing HDX rate.

VII. 12/10- and 12-Helix ^{1}H-NMR Comparisons

The ^{1}H-NMR resonance assignments of the amide protons of 12/10-helical α/γ(**I**)-peptide foldamers reveal an interesting trend: all γ(**I**)-residue amide NH capable of forming 10-atom H-bonds are upfield of the α-residue amide NH capable of forming 12-atom H-bonds. This is suggestive of the γ(**I**) NHs forming less favorable H-bonds than the α NHs. Other data—crystal structures, DMSO titration, and HDX—give us some confidence in interpreting the ^{1}H-NMR data in this way. A few questions we can next ask is: Is this trend common among α/γ-peptides? Is this trend common only among α/γ-peptides which adopt 12/10-helices? Or is this only observed in the 12/10-helical α/γ(**I**)-peptides? **Table S16** shows amide chemical shift data for representative 12/10-helical α/γ-peptides reported in the literature. Also shown, for comparison, is a 12-helical α/γ-peptide. **Figure S11** depicts the data graphically.

Table S16. Amide chemical shift comparison of α/γ-peptides



Compound	γ-Residue	Helix Type	δ[α]1 (ppm)	δ[γ]2 (ppm)	δ[α]3 (ppm)	δ[γ]4 (ppm)	δ[α]5 (ppm)	δ[γ]6 (ppm)	δ[α]7 (ppm)	δ[γ]8 (ppm)
5	γ(**I**)	12/10	5.09	6.69	7.77	6.40	8.52	6.14	8.20	5.90
S12	γ(**I**)	12/10	4.94	7.08	7.79	6.78	8.70	6.74	7.83	6.09
S17[S19]	APCH	12/10	4.99	8.23	8.60	8.00	8.77	5.67	–	–
S18[S21]	AMCP	12/10	5.11	7.47	7.95	7.71	8.72	7.76	8.36	6.61
S19[S1]	γ-Caa	12/10	5.21	7.64	8.03	7.76	8.57	6.90	8.23	–
S20[S22]	γ(**II**)	12	–	5.08	6.62	7.08	7.80	8.15	8.07	–

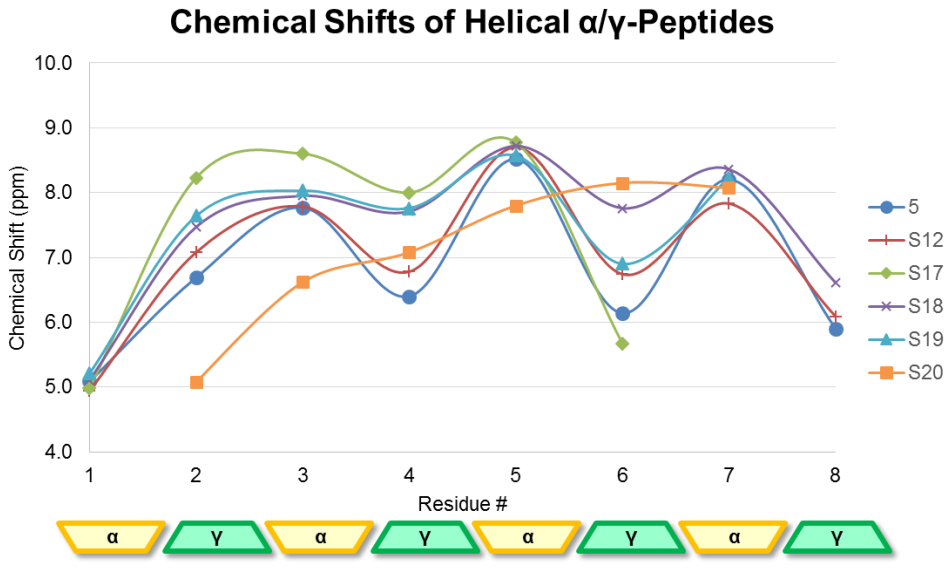


Figure S11. Amide chemical shift data for several 12/10- and 12-helical α/γ-peptides. Lines connecting data points serve as a visual guide.

Immediately apparent from **Figure S11** is the aforementioned trend from the α/γ(**I**)-peptides **5** and **S12** of high α-NH chemical shift/lower γ-NH chemical shift. Compound **S17**, containing γ(**III**) which is an isomer of γ(**I**) with a cyclic constraint at C_α–C_β instead of at C_β–C_γ, features highly downfield chemical shifts of all NH and a smaller difference between α-NH and γ-NH. Compounds **S18** and **S19** behave similarly, with relatively shallow differences between α-NH and γ-NH, although **S19** features a quite upfield [γ]6-NH. Foldamer **S20**, which was found to be 12-helical in solution, does not show the clear chemical shift trend of the other peptides.

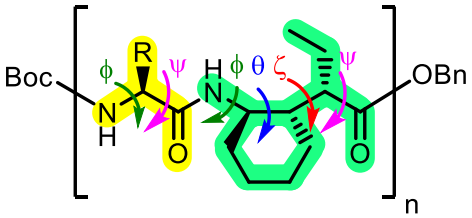
The amide chemical shift trends seem to suggest four features of α/γ-peptide 12/10-helix structure: 1) α/γ-Peptide 12/10-helices generally feature less favorable 10-atom H-bonds donated by γ-NH compared with the 12-atom H-bonds donated by α-NH; 2) A weaker 10-atom/stronger 12-atom H-bond trend manifests most strongly in α/γ(**I**)-peptides; and 3) This trend becomes less distinguishable when the cyclic constraint of γ(**I**) at C_β–C_γ is positioned instead at C_α–C_β by substituting γ(**I**) with γ(**III**). The second and third claims are bolstered by the ease with which amphiprotic solvent-interpolated H-bonds appeared in α/γ(**I**)-peptide 12/10-helix crystals and the lack of this phenomenon in the few other 12/10-helix crystals.

VIII. Helix Structural Parameters

Backbone torsion angle analysis

Below in **Table S16** are listed the backbone torsion angles of the seven described α/γ(**I**)-peptide crystal structures, measured using PyMOL. L-α/(*S*,*S*,*R*)γ(**I**)-peptides are shown for the racemically-grown crystals, and, for ease of comparison, dihedrals of the enantiomers of crystals **1b** and **1c**, which were D-α/(*R*,*R*,*S*)γ(**I**)-peptides, are listed. Compound **2b** crystallized with two conformers in the asymmetric unit, which differ primarily by the benzyl ester O–CH_2 bond torsion angle. Torsion angles within the helical structure vary only subtly; the main text describes conformer **A**.

Table S16. Backbone torsion angles for α/γ(**I**)-peptide crystal structures.[a]



Structure	Residue	ϕ	θ	ζ	ψ
gellman155 **L-1a**	[Phe]1	-61.0			141.6
	[γ(I)]2	131.3	-51.4	107.5	-108.7
	[Phe]3	-78.2			148.0
	[γ(I)]4	124.2	-52.6	165.1	93.6
gellman126 *ent*-**1b**	[Ala]1	-76.4			-20.0
	[γ(I)]2	-57.0	-52.0	133.5	-103.7
	[Ala]3	-69.3			-33.7
	[γ(I)]4	101.7	-54.9	157.6	-81.8
gellman124a *ent*-**1c**	[Ala]1	-64.4			152.5
	[γ(I)]2	99.4	-58.9	150.3	-116.0
	[Ala]3	-72.7			124.7
	[γ(I)]4	125.3	-55.4	169.3	-63.9

	[Val]1	-60.9			137.6
	[γ(I)]2	149.5	-50.7	91.0	-122.1
gellman149	[Val]3	-55.4			138.6
2a	[γ(I)]4	99.3	-61.6	156.4	-104.3
	[Val]5	-136.4			165.0
	[Ala]1	-68.8			148.7
	[γ(I)]2	107.1	-59.9	141.9	-111.6
gellman153	[Ala]3	-69.4			144.0
2b (A)	[γ(I)]4	99.6	-62.6	154.2	-110.6
	[Ala]5	-69.8			162.4
	[Ala]1	-67.1			148.2
	[γ(I)]2	109.6	-60.6	141.2	-107.5
gellman153	[Ala]3	-62.8			146.8
2b (B)	[γ(I)]4	97.9	-58.5	150.4	-115.3
	[Ala]5	-70.8			156.1
	[Phe]1	-55.2			136.2
	[γ(I)]2	140.5	-45.7	87.6	-124.7
gellman157	[Phe]3	-54.9			144.7
L-3a	[γ(I)]4	126.2	-57.2	119.8	-114.1
	[Phe]5	-104.6			137.5
	[γ(I)]6	123.5	-51.2	176.5	-59.9
	[Phe]1	-63.9			144.1
	[γ(I)]2	139.5	-50.3	96.8	-116.6
gellman156	[Phe]3	-60.8			148.0
L-3b	[γ(I)]4	116.3	-62.9	141.1	-110.2
	[Phe]5	-79.4			129.1
	[γ(I)]6	127.4	-50.0	178.7	115.3
α Median		-68.8			144.1
α Std Dev		18.4			52.6
α 12/10-Helix[a] Median		-61.9			144.4
α 12/10-Helix[a] Std Dev		6.9			4.4
γ(I) Median		119.9	-55.2	146.1	-109.4
γ(I) Std Dev		43.1	5.1	27.7	68.4
γ(I) 12/10-Helix[a] Median		121.2	-57.8	130.4	-112.8
γ(I) 12/10-Helix[a] Std Dev		18.6	5.9	26.4	6.2

[a]This dihedral is: 1) present in a 12/10-helical crystal structure, and 2) not a C-terminal residue.

A few trends can be identified from analyzing the torsion angles in the crystal structures. First, C-terminal residues of 12/10-helical foldamers adopt more varied torsions than the residues at the N-terminus or internal in the sequence. This may reflect either the better ability of these residues to adopt their more highly preferred torsion angles or the perturbation required to form intermolecular H-bonds with lattice neighbors.

The most intriguing find of the crystal structure set is the unusual ambivalence these foldamers appear to display in formation of the 10-atom H-bond. With that in mind, an important question to ask is: Which torsions distinguish "closed" 10-atom H-bonds—those that form intramolecular 10-atom H-bonds fitting conventional geometric criteria—from those within "opened" 10-atom H-bonds—those that are able to form 10-atom H-bonds, but do not?

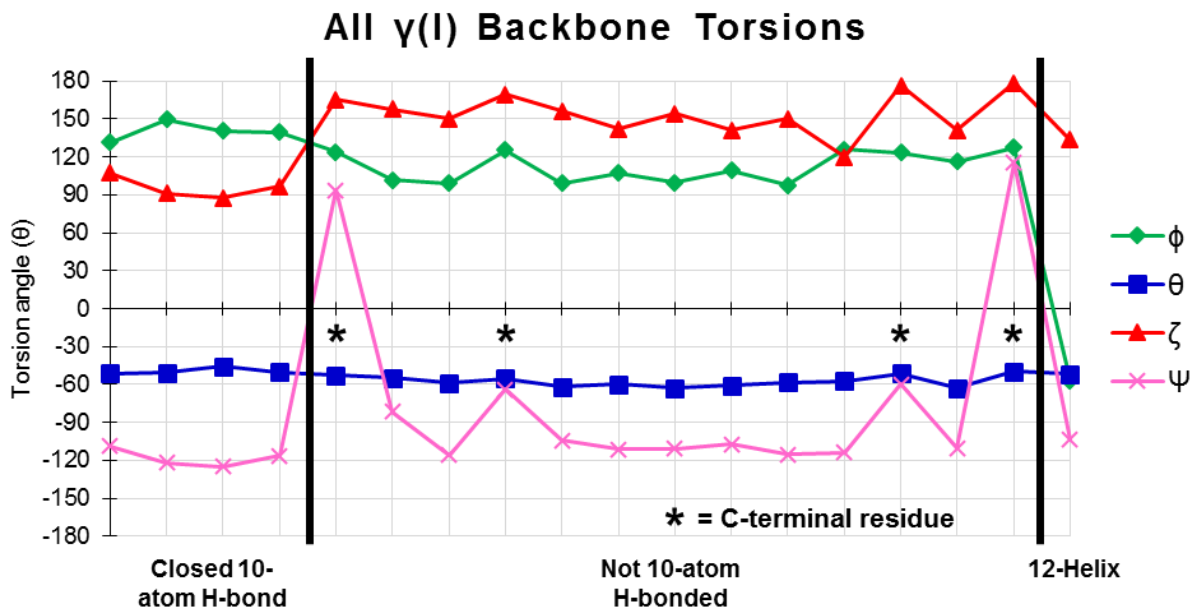


Figure S12. Torsion angles adopted by γ(**I**) residues in crystal structures reported in the main text. Torsion angles of the enantiomers of **1b** and **1c** are shown. Each value on the x-axis represents a different γ(**I**) residue. Residues with torsion angles within a closed 10-atom H-bonds are shown at the left, followed after the vertical bar by residues with torsion angles not within 10-atom H-bonds (this includes solvent-bridged, poor H-bond geometry, intermolecularly H-bonded, or not H-bonded), followed by the one 12-helical residue in tetramer **1b**. Asterisks signify γ(**I**) residues at the C-terminus, which cannot form 10-atom H-bonds.

The 12/10-helix α residue torsion angles cluster relatively tightly around [ϕ,ψ] = [-62,144], in the β region of the Ramachandran plot. These torsions do not vary much between among α residues with torsions within closed or within opened 10-atom H-bonds. Most interesting are the γ(**I**) torsion angles (**Figure S12**). Expectedly, θ, the torsion angle about the constrained C_β–C_γ backbone cyclohexyl bond, does not vary much no matter the structural context of the residue. Torsion ψ in γ(**I**) residues also adopts similar values in all structures, excluding the C-terminal residues in which ψ defines the torsion angle about the C_α–C_{CO}, the definition of which includes the ester oxygen atom.

The torsion angles ϕ about the N–C_γ bond and ζ about the C_α–C_β bond differ the most between the γ(**I**) residues within a closed 10-atom H-bond and those within an opened H-bond. **Figure S13** below depicts how these torsions result in differing structures, focusing here on **3b** which contains both a closed 10-atom H-bond and an opened 10-atom H-bond. The ϕ of the γ(**I**) residue within the closed 10-atom H-bond (140.5°) is greater than the ϕ of the γ(**I**) within the opened 10-atom H-bond (126.2°), and the ζ within the closed 10-atom H-bond (87.6°) is smaller than the ζ of the opened 10-atom H-bond (119.8°). The summed effect of these two torsional shifts is to reposition the 10-atom H-bond acceptor carbonyl oxygen atom farther from the amide H-bond donor hydrogen atom. Notably, the γ(**I**) residue within this opened 10-atom H-bond is also within a closed 12-atom H-bond. The torsion perturbation resulting in the opening of the 10-atom H-bond thus does not result in disruption of the 12-atom H-bond.

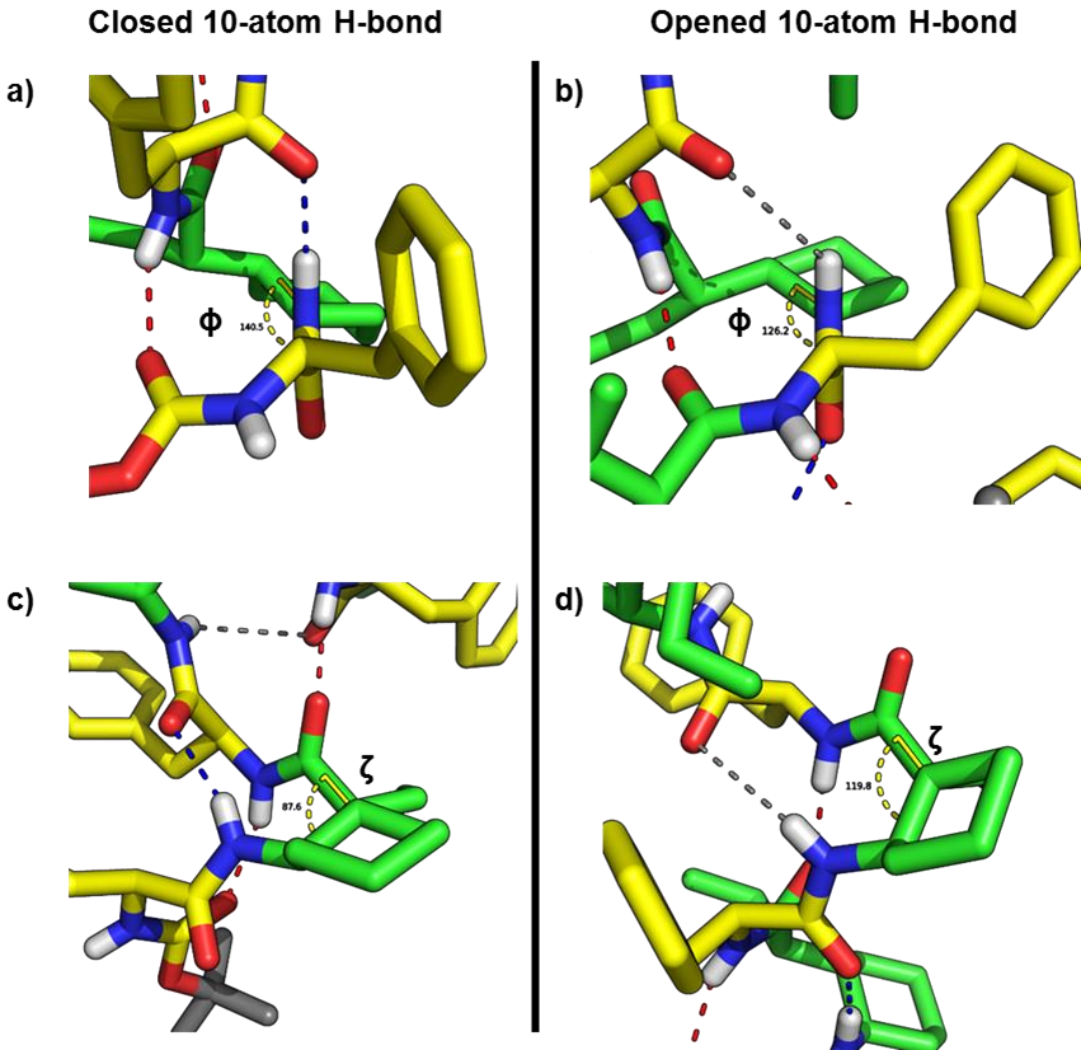


Figure S13. Comparison of torsion angles ϕ and ζ within structure **3b**. Top: comparison of typical ϕ for **a)** 12/10-helical γ(**I**) of 140.5° and **b)** non-12/10-helical γ(**I**) of 126.2°. Bottom: comparison of typical ζ for **c)** 12/10-helical γ(**I**) of 87.6° and **d)** non-12/10-helical γ(**I**) of 119.8°. The closed 10-atom H-bond is depicted with blue dashes, and the opened 10-atom H-bond is depicted with gray dashes. Note the closed 12-atom H-bond in red, formed in both structures.

H-Bond Parameters

Table S17 lists the H-bond distances and angles derived from the crystal structure data. H-bond donors and acceptors are listed as (Xxx,y) where Xxx = residue type of the donor/acceptor and Y = position in the sequence of the donor/acceptor (Boc = position 0). H-bond distances are listed as the donor-acceptor distance (D···A). Bond angles were measured with uncorrected polar hydrogen atom positions. H-bond size is undefined for intermolecular H-bonds, and angle C=O···H is undefined for non-carbonyl H-bond acceptors. The two conformers of compound **2b** feature slightly different geometric H-bond parameters for the C-terminal 12-atom H-bond, although both foldamers had an EtOH molecule in the same position. Conformer **A** is discussed in the main text.

Table S17. Intramolecular H-Bond Parameters for Crystal Structures

Compound	H-Bond Type	H-Bond Size	D···A (Å)	C=O···H (°)	A···H−D (°)
1a	C=O(Boc,0)→HN(Phe,3)	12-atom	2.83	144.2	170.4
	C=O(Phe,3)→HN(γ,2)	10-atom	3.00	127.7	171.3
1b	C=O(Boc,0)→HN(Ala,3)	12-atom	3.06	134.7	162.6
	C=O(Ala,1)→HN(γ,4)	12-atom	3.00	127.9	161.2
1c	C=O(Boc,0)→HN(Ala,3)	12-atom	3.10	158.4	170.3
	C=O(Ala,3')→HN(γ,2)	−	2.91	142.5	177.3
	C=O(Ala,3)→HOH	−	2.86	123.8	162.1
2a	C=O(Boc,0)→HN(Val,3)	12-atom	3.02	144.0	168.2
	C=O(Val,3)→HN(γ,2)	10-atom	3.15	121.4	163.6
	C=O(γ,2)→HN(Val,5)	12-atom	3.11	156.5	164.1
2b (A)	C=O(γ,i)→HN(Ala,i+3)	12-atom	2.91	166.0	165.8
	EtHO→HN(γ,2)	−	2.95	−	148.0
	C=O(Ala,3)→HOEt	−	2.73	132.7	160.2
	C=O(γ,2)→HN(Ala,5)	12-atom	2.95	158.6	173.1
2b (B)	C=O(γ,i)→HN(Ala,i+3)	12-atom	2.93	160.7	167.5
	EtHO→HN(γ,2)	−	2.89	−	145.1
	C=O(Ala,3)→HOEt	−	2.70	138.4	166.8
	C=O(γ,2)→HN(Ala,5)	12-atom	2.98	144.8	154.4
3a	C=O(Boc,0)→HN(Phe,3)	12-atom	2.86	145.2	155.5
	C=O(Phe,3)→HN(γ,2)	10-atom	3.02	126.5	162.6
	C=O(2,i)→HN(Phe,5)	12-atom	2.85	149.9	171.9
	C=O(Phe,5)→HN(γ,4)	10-atom	4.09	110.1	135.0
3b	C=O(Boc,0)→HN(Phe,3)	12-atom	2.92	149.4	159.4
	C=O(Phe,3)→HN(γ,2)	10-atom	2.99	129.9	169.7
	C=O(γ,2)→HN(Phe,5)	12-atom	2.90	172.4	167.3
	H_2O→HN(γ,4)	−	2.86	−	156.2
	C=O(Phe,5)→HOH	−	2.78	129.7	158.9
		10-atom averages	3.04	126.4	166.8
		12-atom averages	2.96	1150.9	165.1

[a]Although parameters are listed here, these atoms do not fit conventional geometric criteria for a hydrogen bonding interaction and are not included in averages.

12/10-Helix Parameters

Below in **Table S18** are shown the helix parameters of α/γ(**I**)-peptide 12/10-helices calculated via reported methods.[S23] All helix parameter values for the 12/10-helix are very close to those of the 12-helix, although the 12/10-helix appears to be slightly narrower in radius. The 12/10- and 12-helices are geometrically very similar despite the difference in H-bonding directionality.

Table S18. Helix parameters for select peptide helix types and for α/γ-peptide 12/10-helices

Backbone	Peptide	Helix type	res/turn	rise/turn (Å)	rise/res (Å)	radius (Å)
α		α-helix[S24]	3.5	5.4	1.5	2.3
α		3_{10}-helix[S24]	3.2	5.8	1.8	2.0
γ	**EtACHA**	14-helix[S25]	2.5	5.5	2.1	2.9
α/γ	**Ala/EtACHA 6mer**	12-helix[S1,a]	2.7	5.5	2.1	2.3
α/γ	**1a**	12/10	2.7	5.8	2.1	2.2
α/γ	**2a**	12/10	2.6	5.4	2.0	2.2
α/γ	**2b**	12/10	2.4	5.5	2.3	1.8
α/γ	**3a**	12/10	2.6	5.5	2.1	2.2
α/γ	**3b**	12/10	2.6	5.5	2.1	2.1
α/γ	**Ala/APCH 4mer**	12/10	2.5	5.3	2.1	1.8
		12/10-Average	**2.6**	**5.5**	**2.1**	**2.1**

[a]Recalculated from ref. using C_β(EtACHA) instead of C_α(EtACHA) as points to define the helix.

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Note: Crystallographic section contains a separate set of references.

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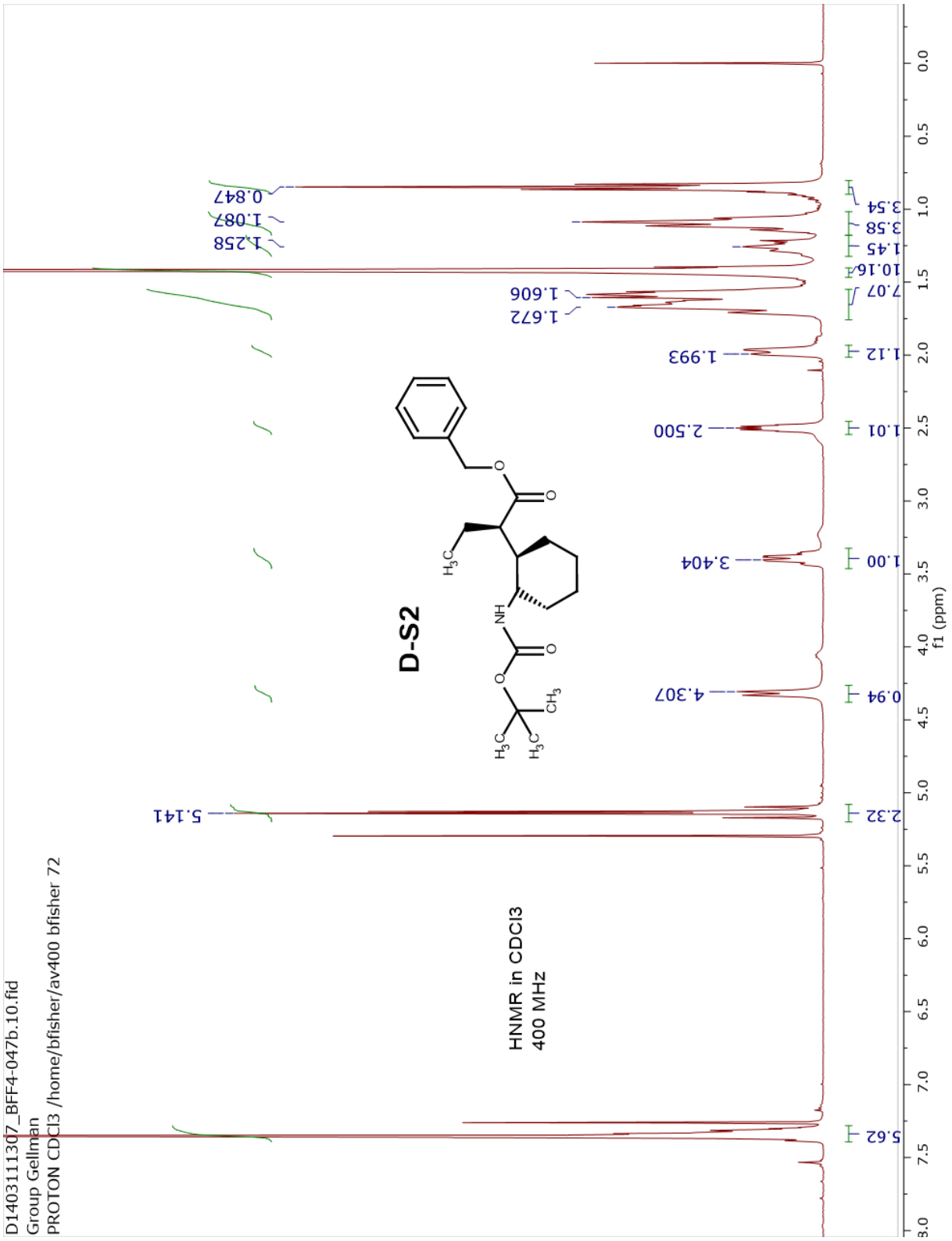
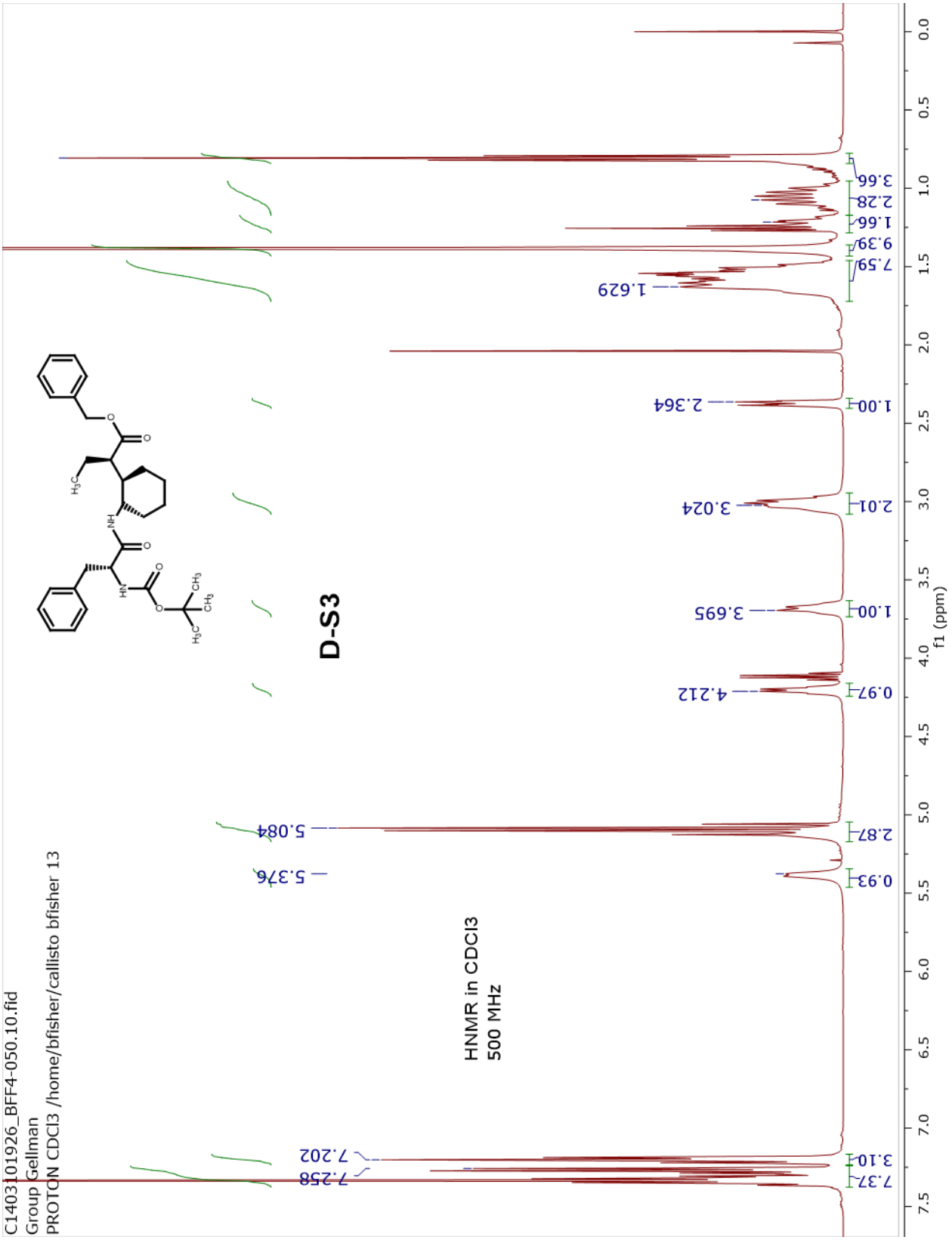
X. NMR Spectra

a. ^{1}H-NMR Spectra

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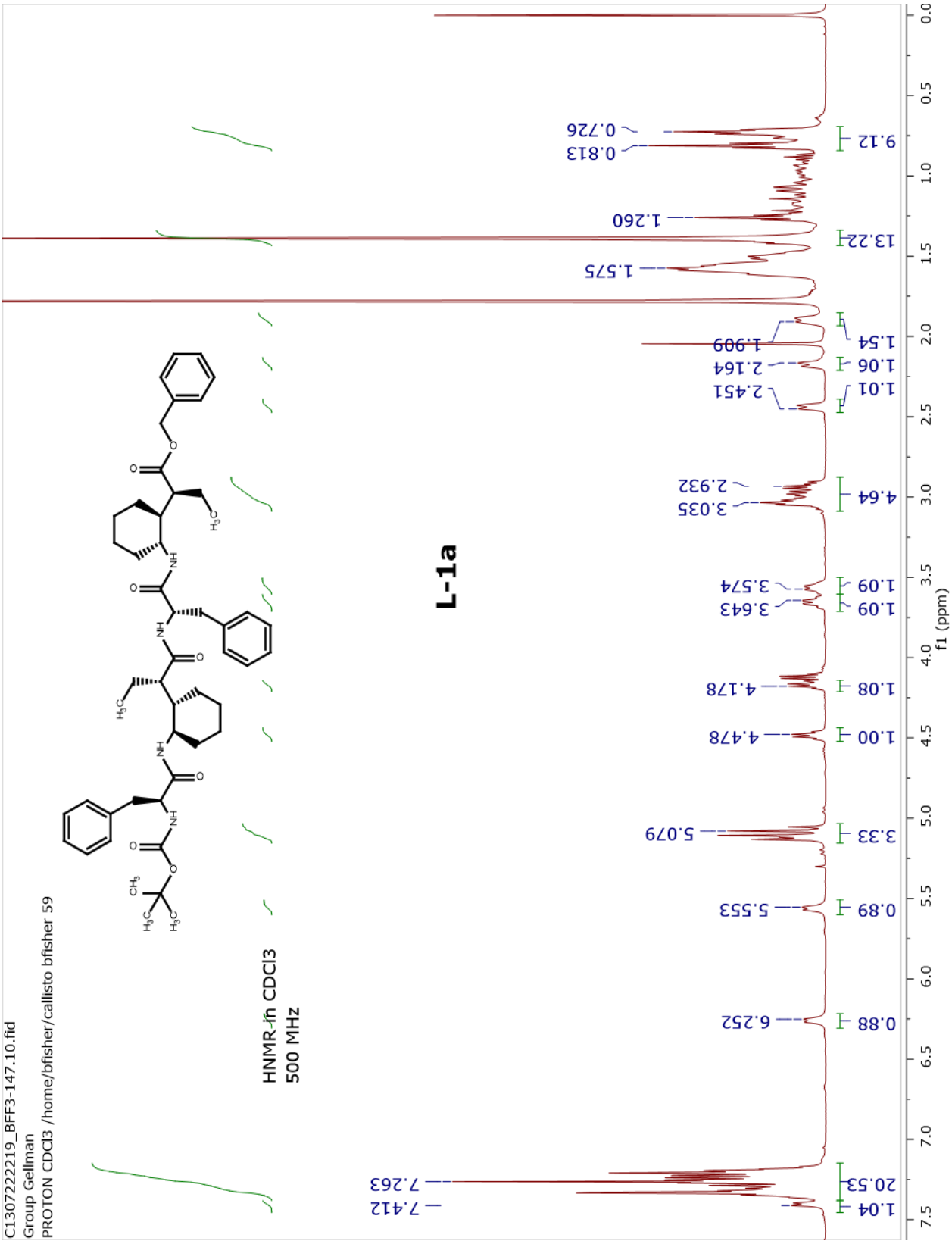
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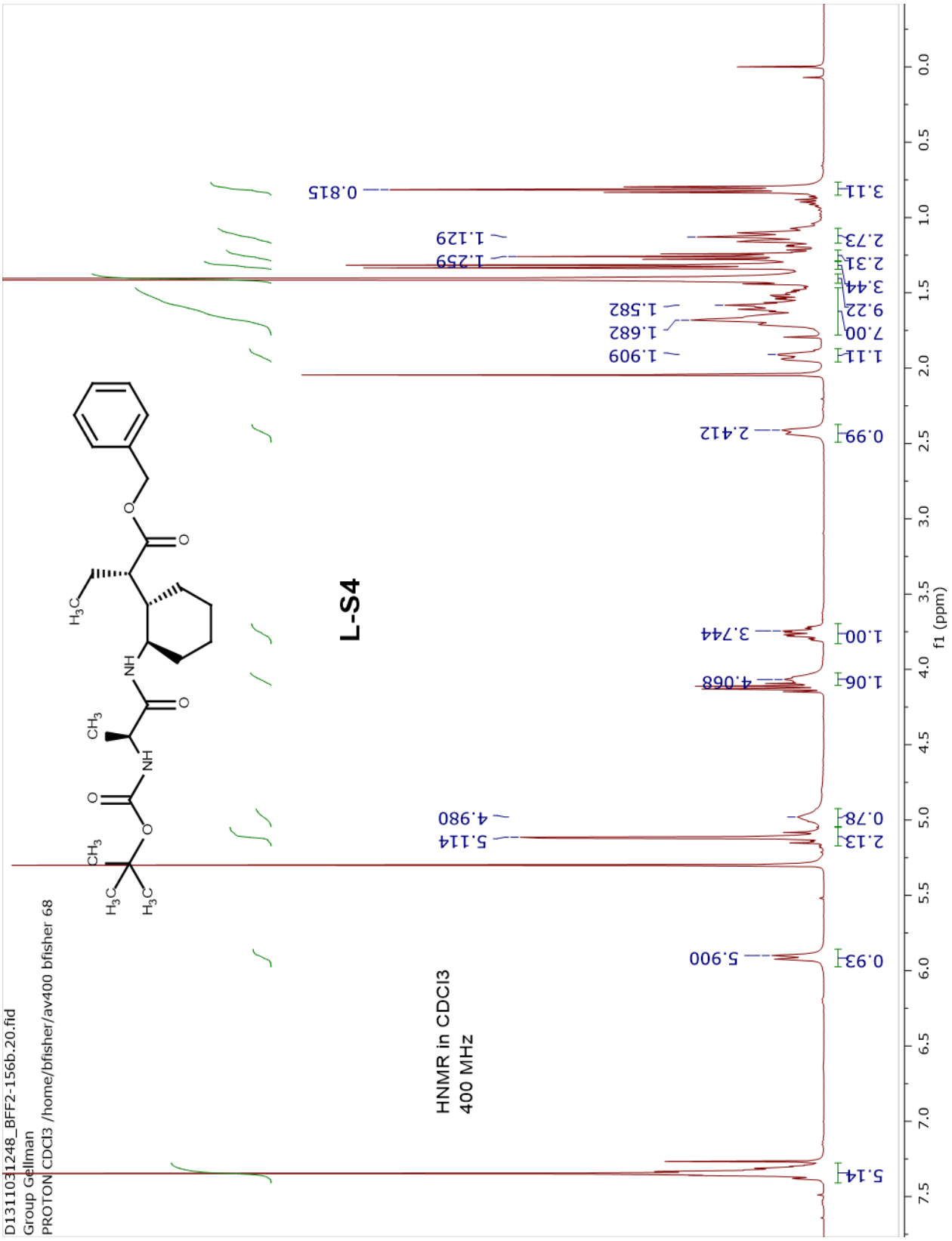
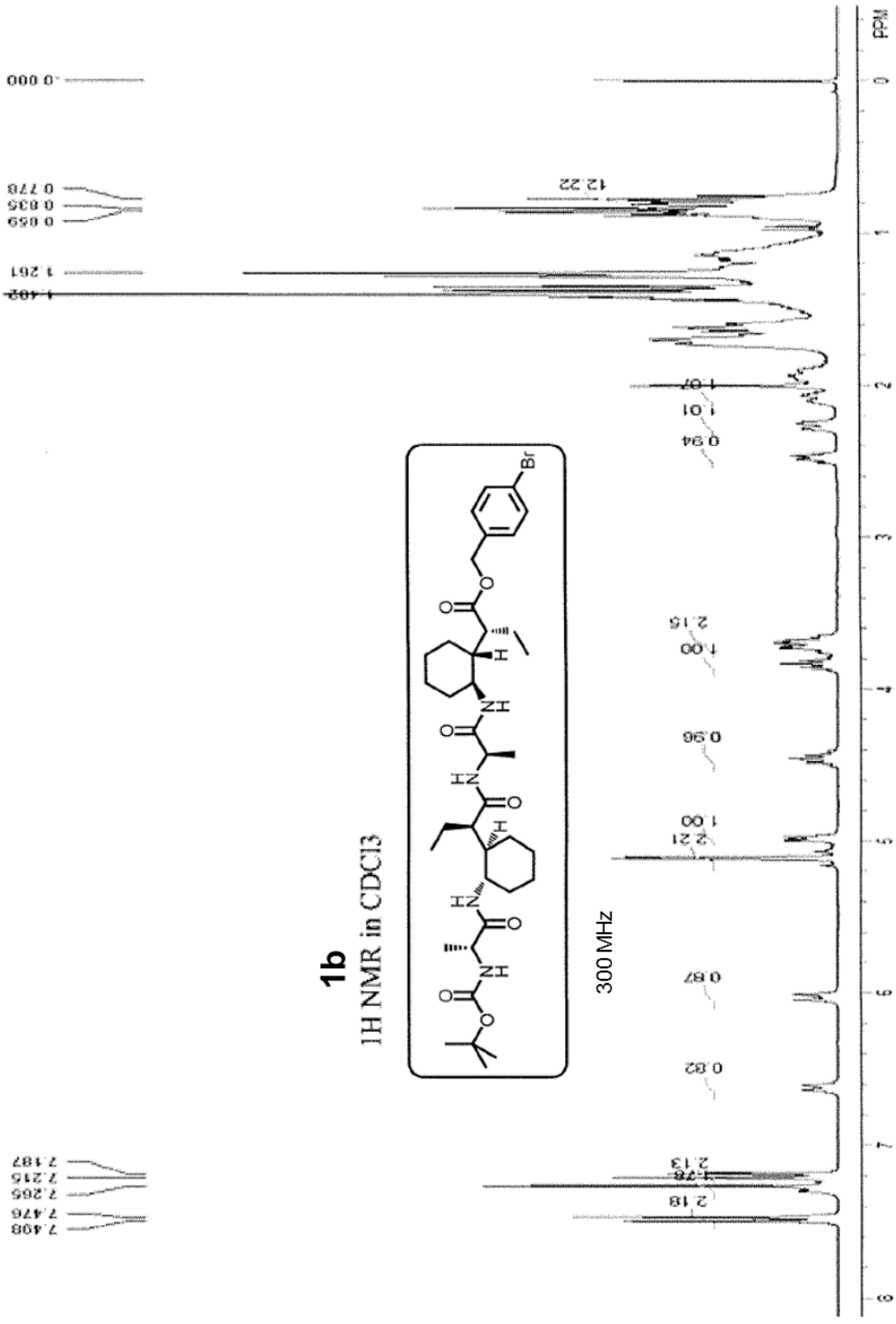
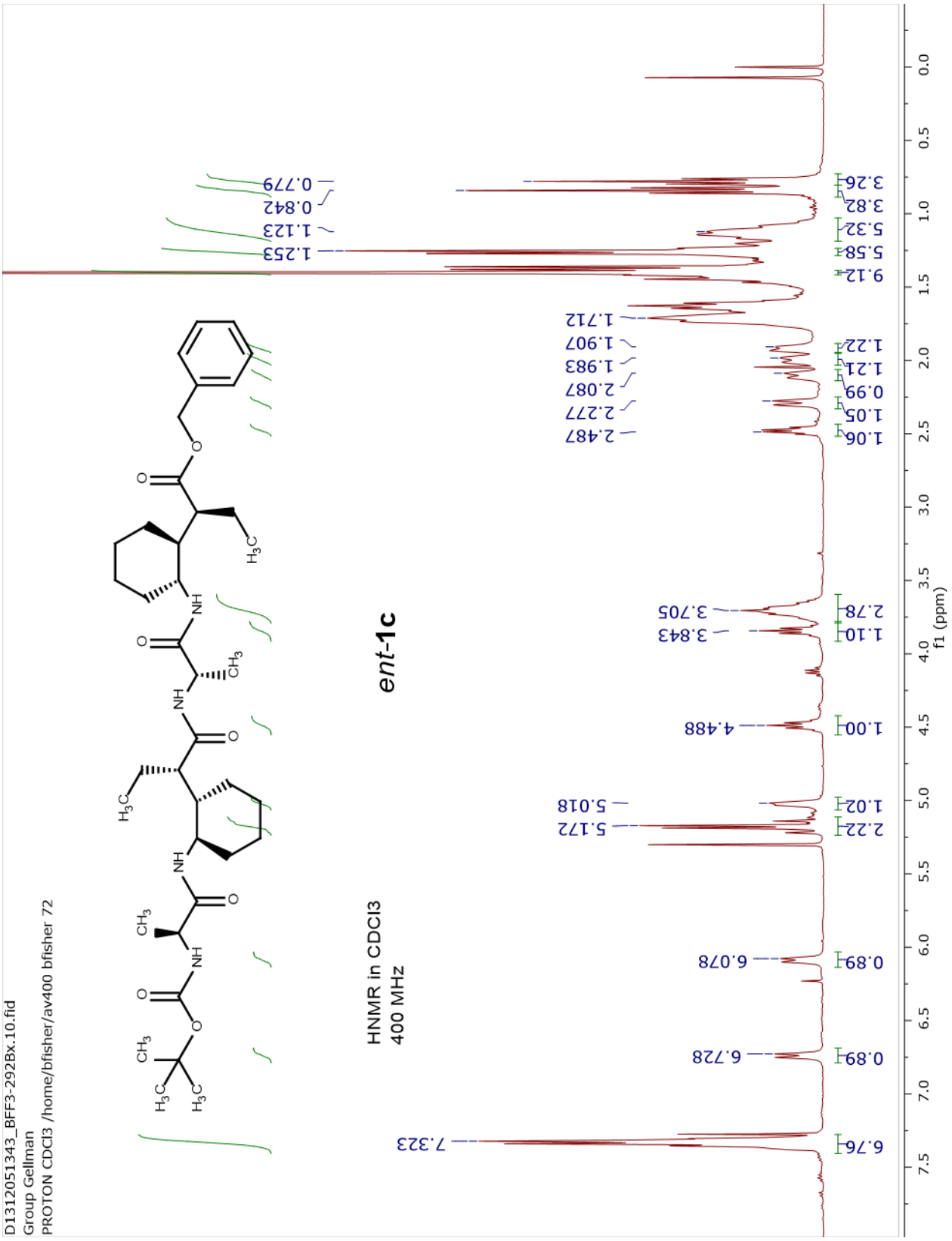
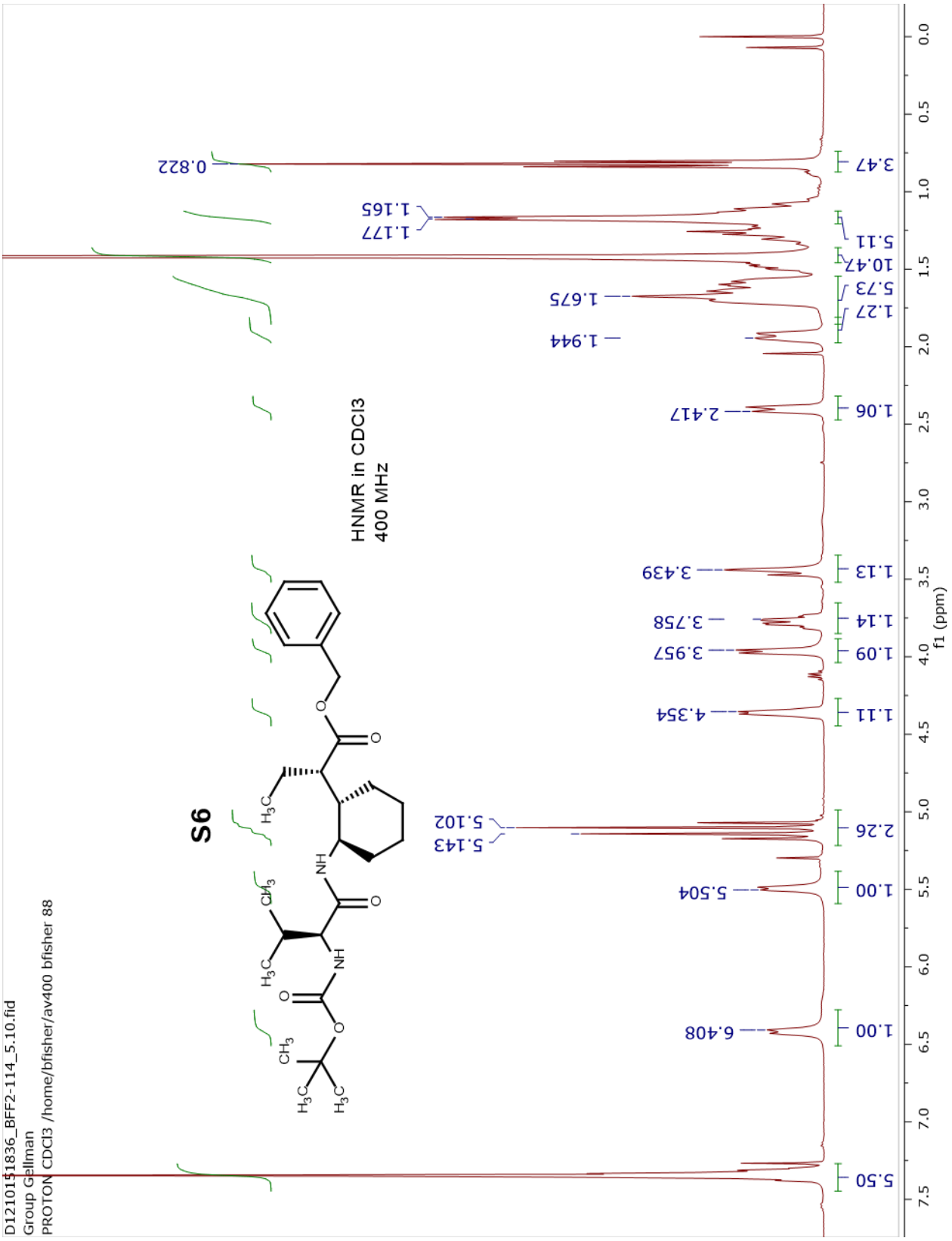
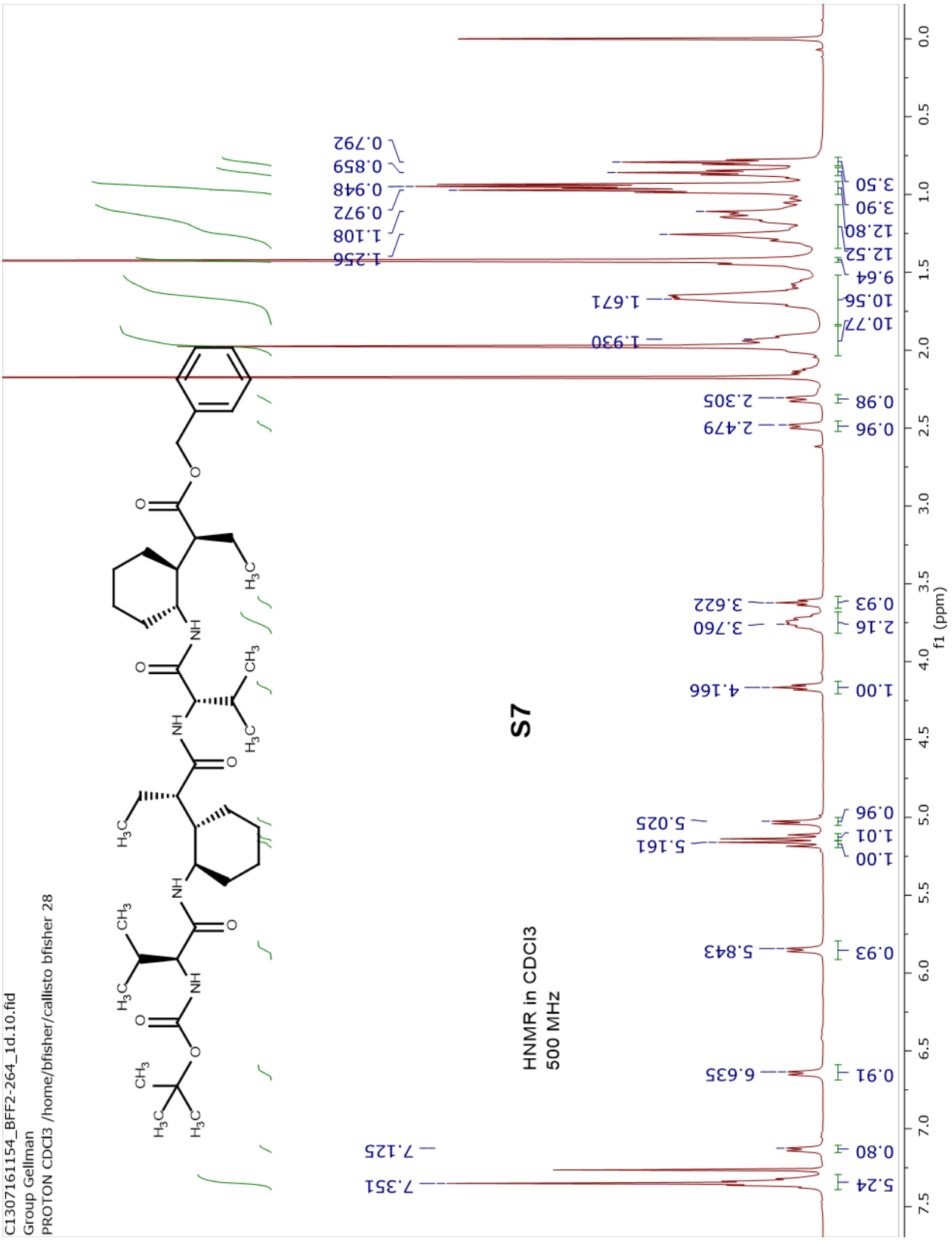
1.01
2.451
1.06
2.164
1.54
1.909
13.22
1.575
1.260
9.12
0.813
0.726
f1 (ppm)

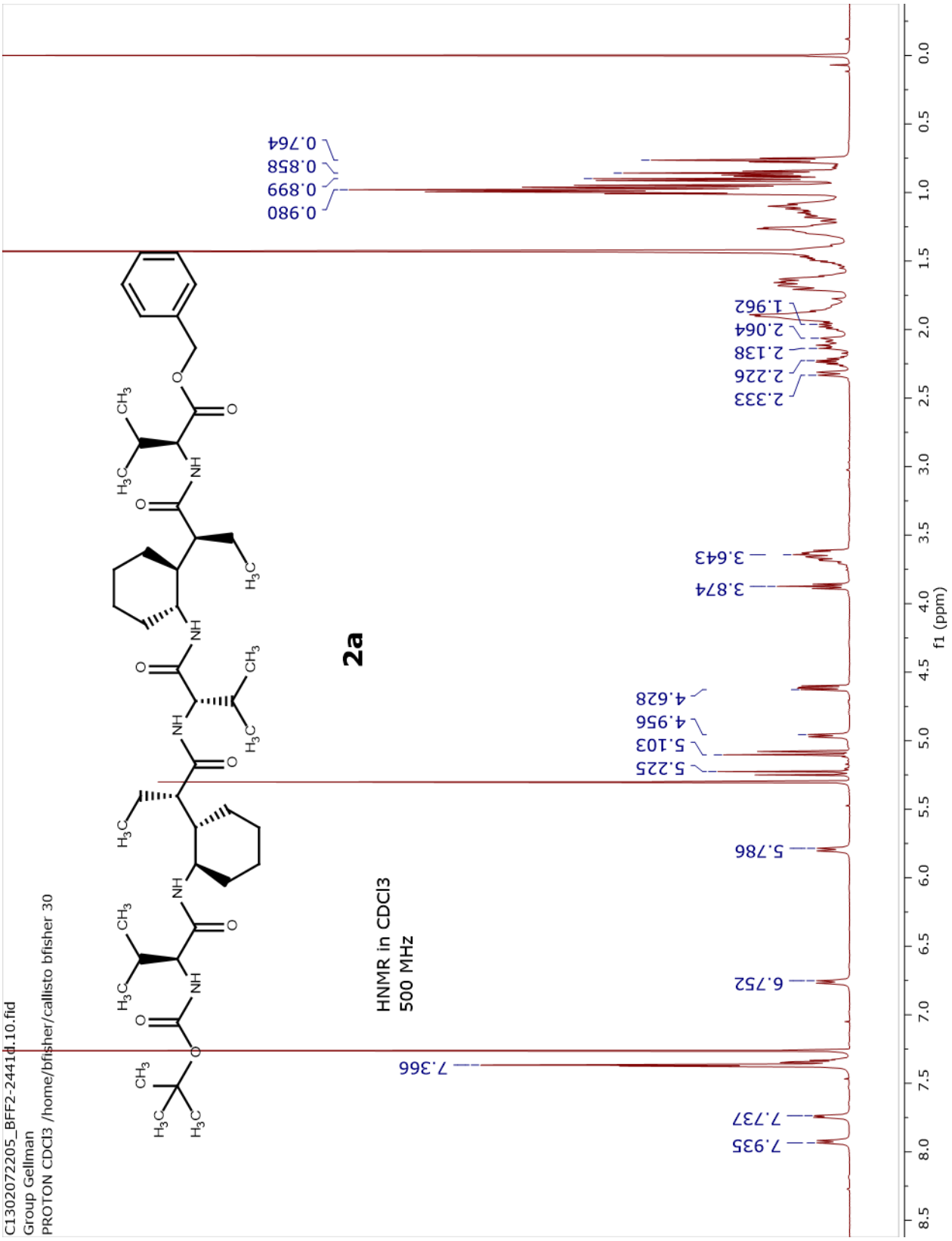

D131103_1248_BFF2-156b.20.fid
Group Gellman
PROTON CDCl3 /home/bfisher/av400 bfisher 68
HNMR in CDCl3
400 MHz
L-S4
0.815
1.129
1.259
1.582
1.682
1.909
2.412
3.744
4.068
4.980
5.114
5.900
3.11
2.73
2.31
3.44
9.22
7.00
1.11
0.99
1.00
1.06
0.78
2.13
0.93
5.14
f1 (ppm)

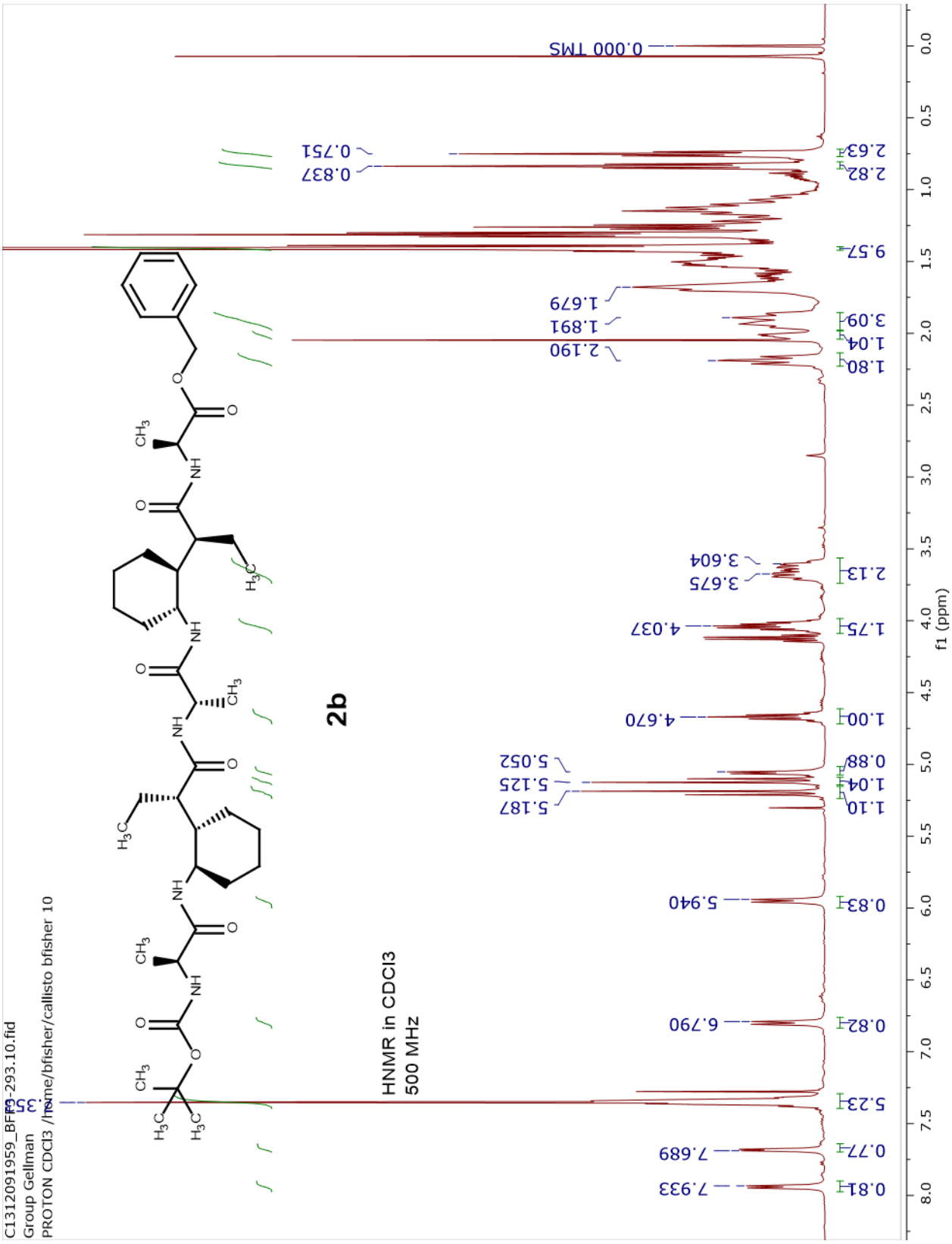

1b
1H NMR in CDCl3
300 MHz
Br
PPM

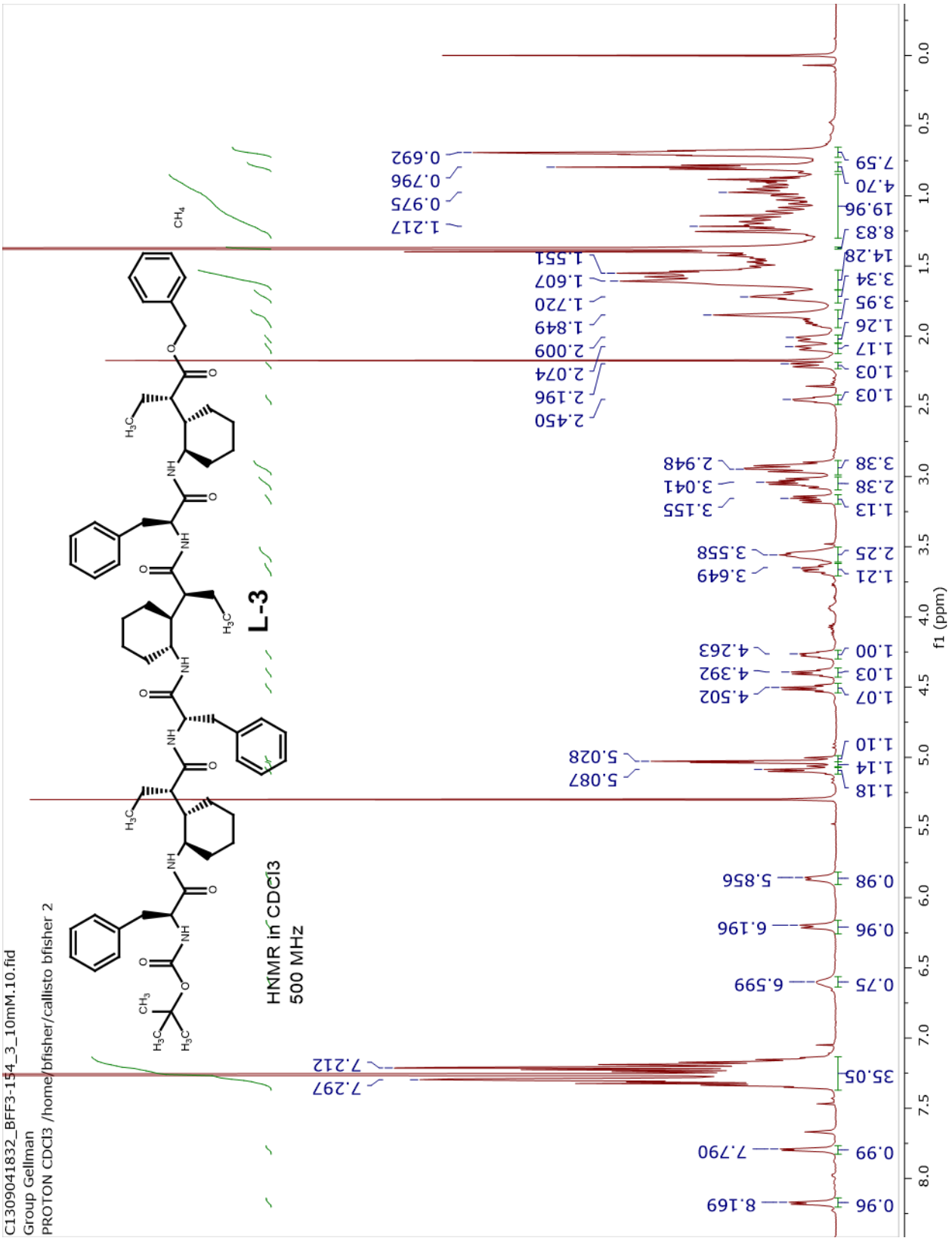

D1312051343_BFF3-292Bx.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/av400 bfisher 72
HNMR in CDCl3
400 MHz
ent-1c
7.323
6.728
6.078
5.172
5.018
4.488
3.843
3.705
2.487
2.277
2.087
1.983
1.907
1.712
1.253
1.123
0.842
0.779
6.76
0.89
0.89
2.22
1.02
1.00
1.10
2.78
1.06
1.05
0.99
1.21
1.22
9.12
5.58
5.32
3.82
3.26
f1 (ppm)

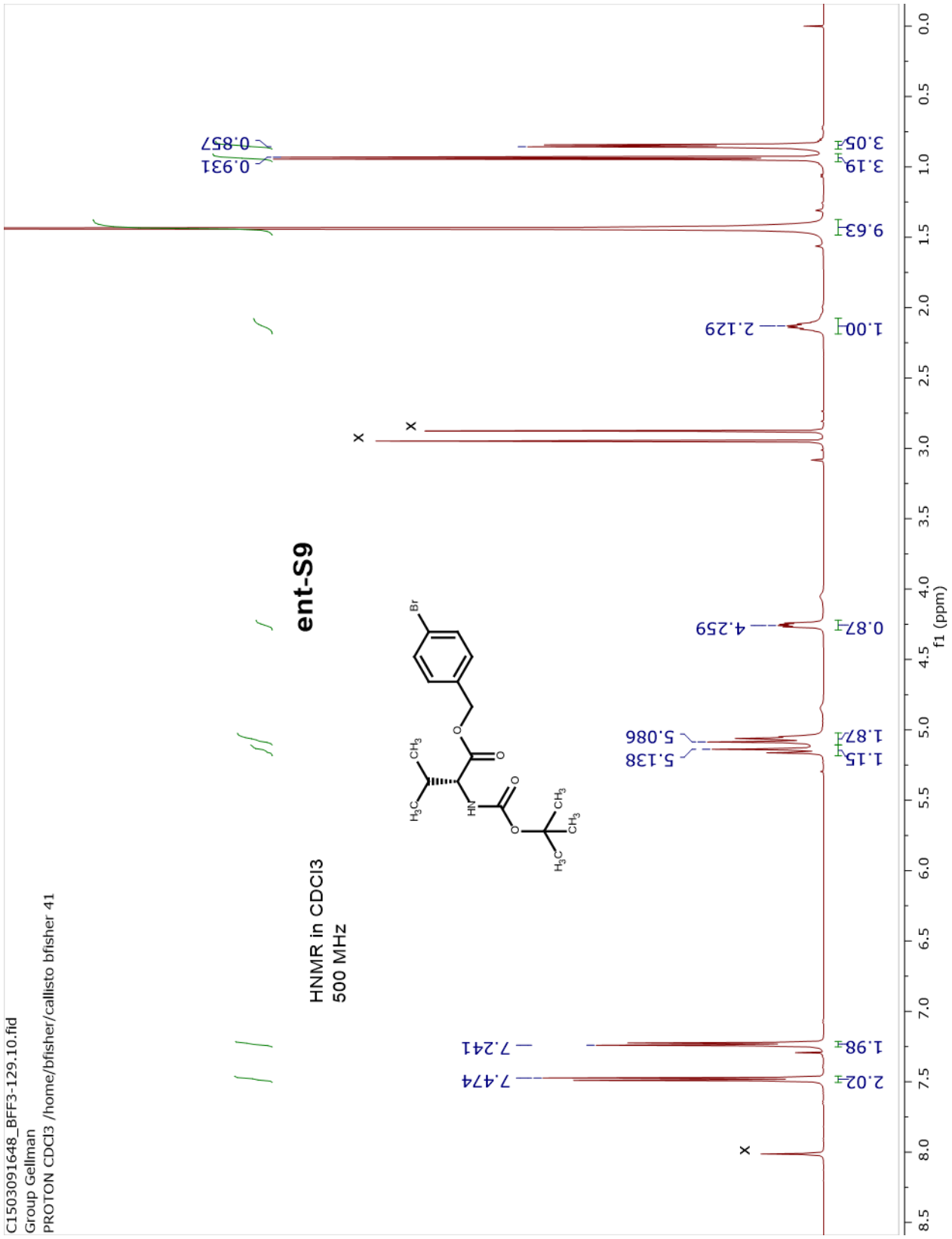

D121015_1836_BFF2-114_5.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/av400 bfisher 88
HNMR in CDCl3
400 MHz
S6
f1 (ppm)
0.822
1.165
1.177
1.675
1.944
2.417
3.439
3.758
3.957
4.354
5.102
5.143
5.504
6.408
5.50
1.00
1.00
2.26
1.11
1.09
1.14
1.13
1.06
1.27
5.73
10.47
5.11
3.47

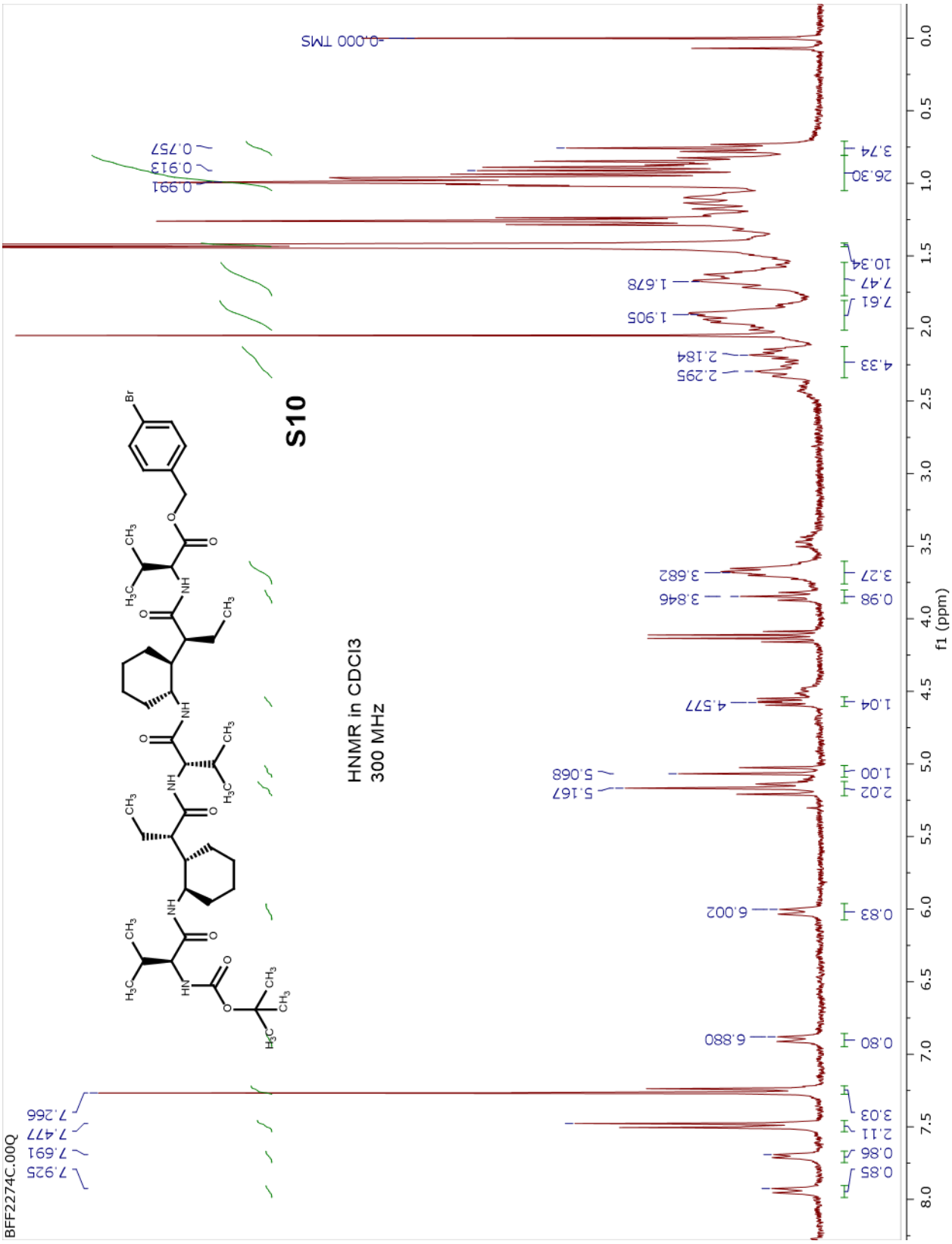

C1307161154_BFF2-264_1d.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 28
HNMR in CDCl3
500 MHz
S7
f1 (ppm)

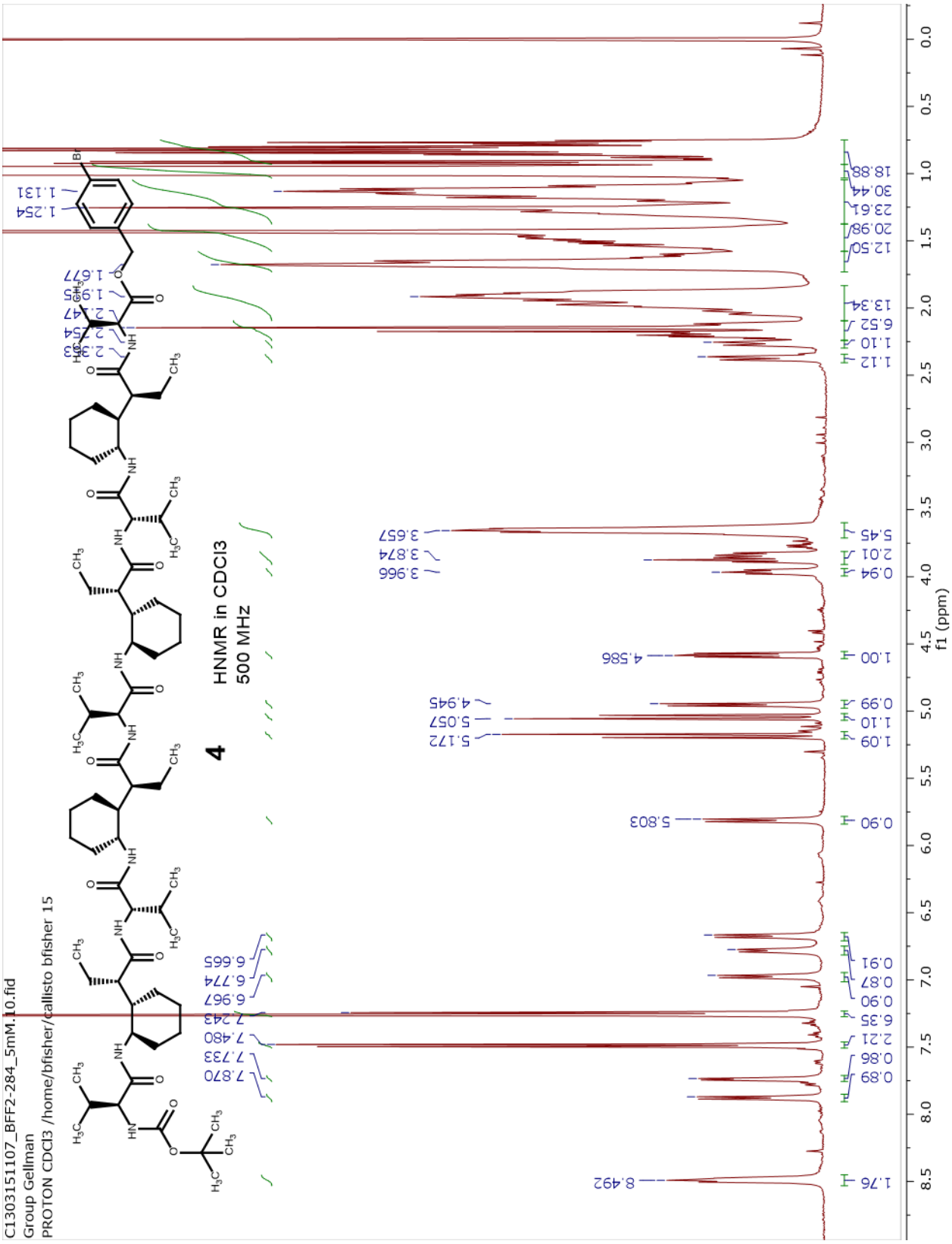

C1302072205_BFF2-2441d.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 30
2a
HNMR in CDCl3
500 MHz
0.764
0.858
0.899
0.980
1.962
2.064
2.138
2.226
2.333
3.643
3.874
4.628
4.956
5.103
5.225
5.786
6.752
7.366
7.737
7.935
f1 (ppm)

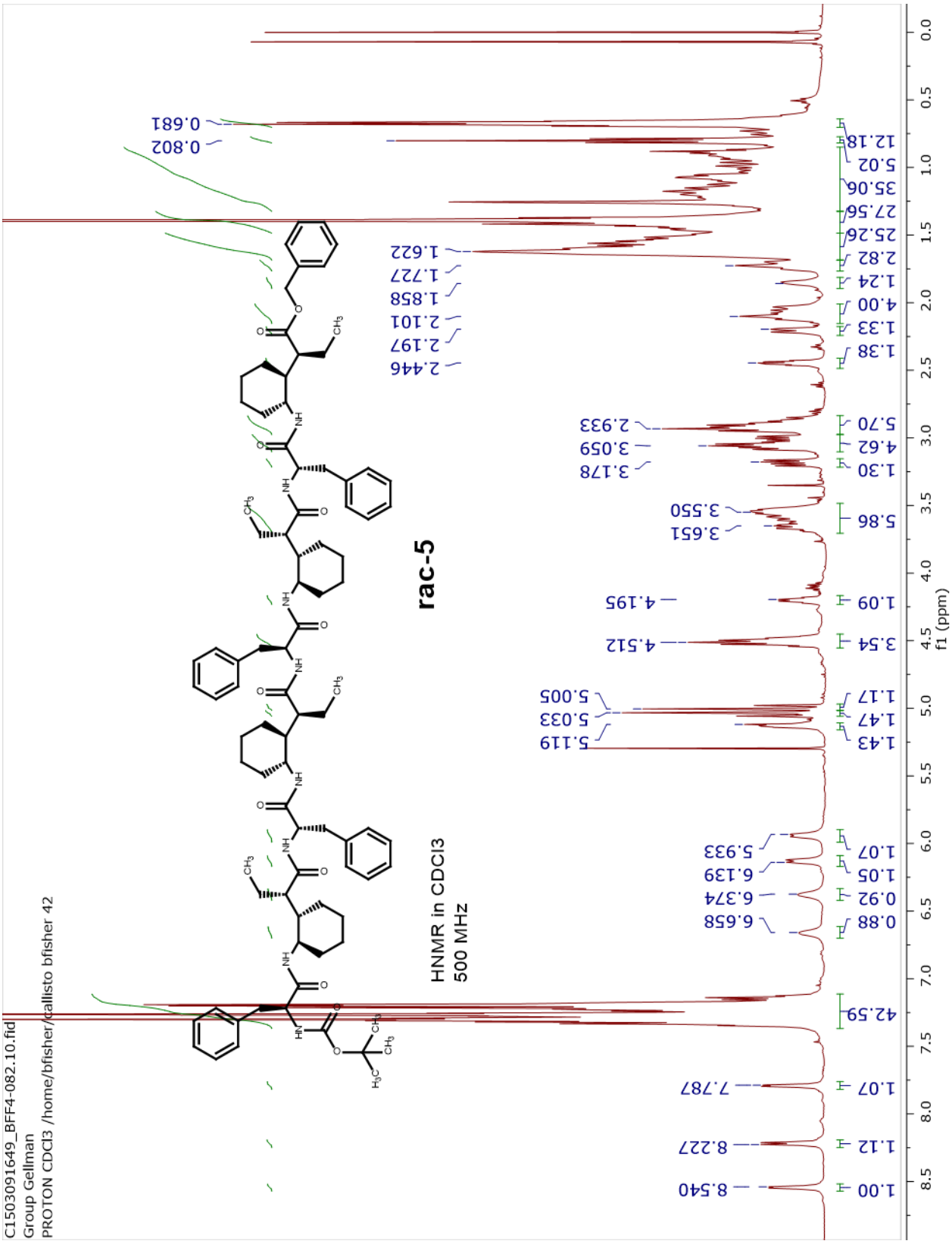

C131209195 9_BFB-293.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 10
HNMR in CDCl3
500 MHz
2b
0.000 TMS
0.751
0.837
1.679
1.891
2.190
3.604
3.675
4.037
4.670
5.052
5.125
5.187
5.940
6.790
7.689
7.933
2.63
2.82
9.57
3.09
1.04
1.80
2.13
1.75
1.00
0.88
1.04
1.10
0.83
0.82
5.23
0.77
0.81
f1 (ppm)

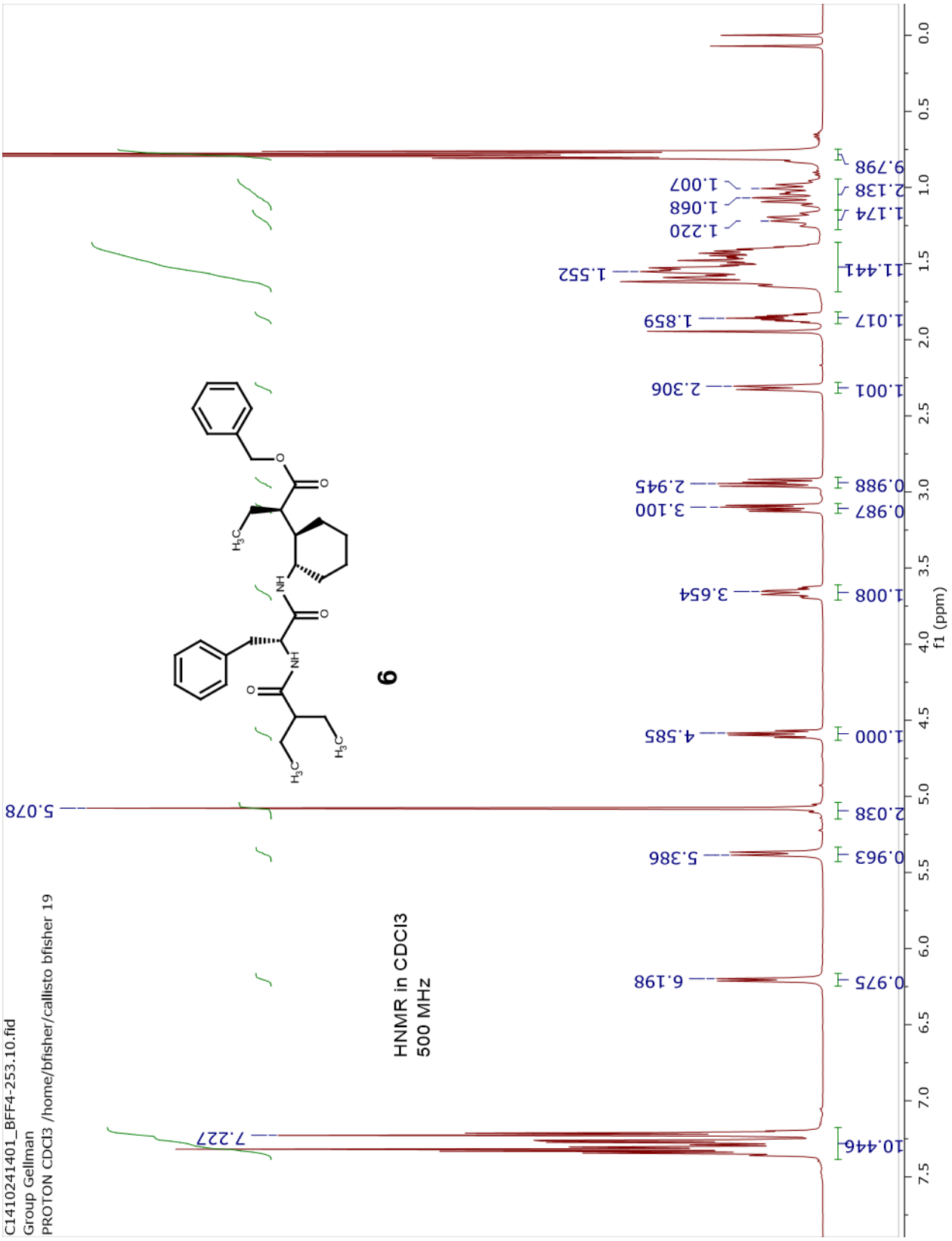

C130904832_BFF3-154_3_10mM.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 2
HNMR in CDCl3
500 MHz
L-3
f1 (ppm)

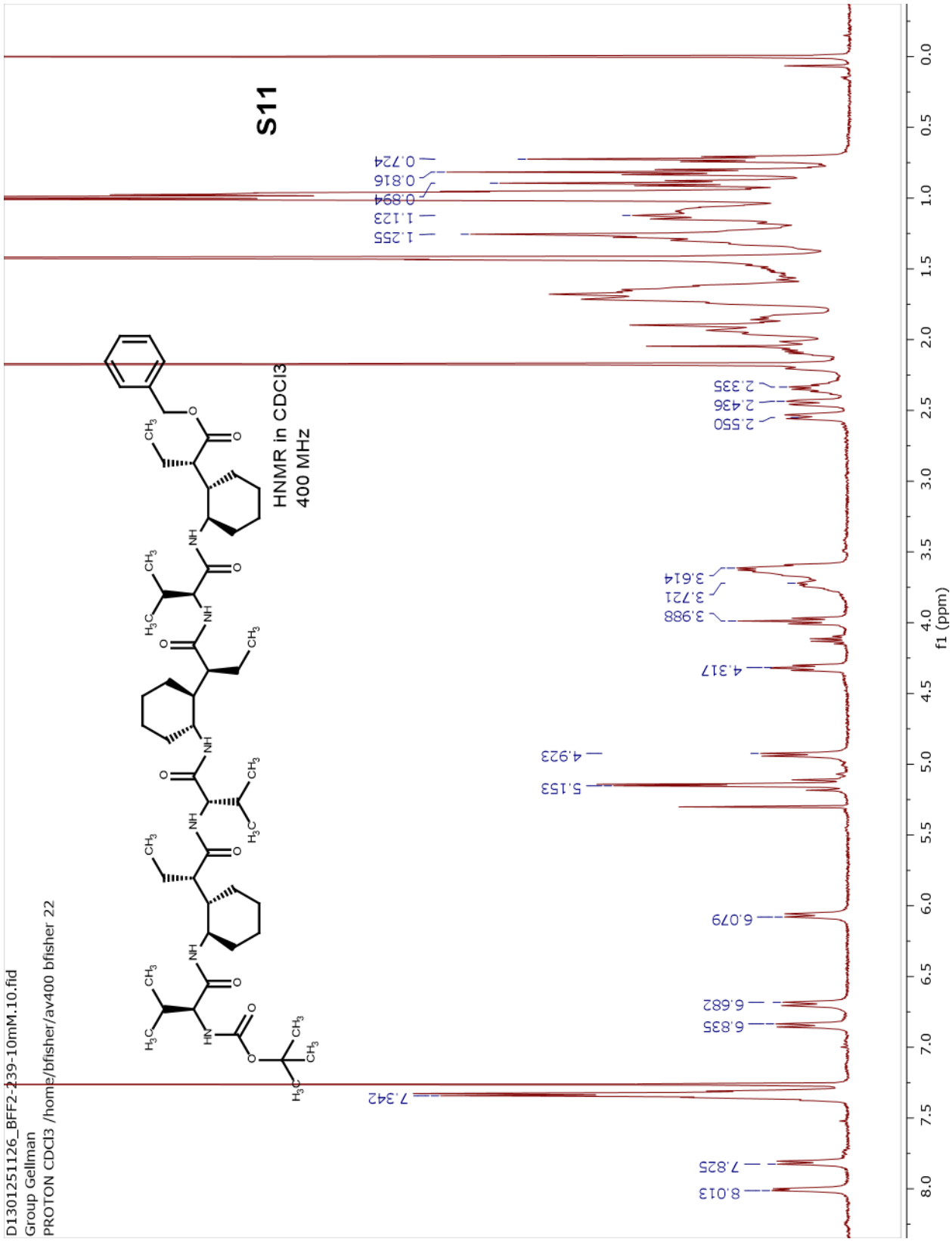

C1503091648_BFF3-129.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 41
HNMR in CDCl3
500 MHz
ent-S9
Br
O
CH3
H3C
O
HN
O
O
CH3
CH3
H3C
x
x
x
0.857
0.931
3.05
3.19
9.63
2.129
1.00
4.259
0.87
5.086
5.138
1.87
1.15
7.241
7.474
1.98
2.02
f1 (ppm)

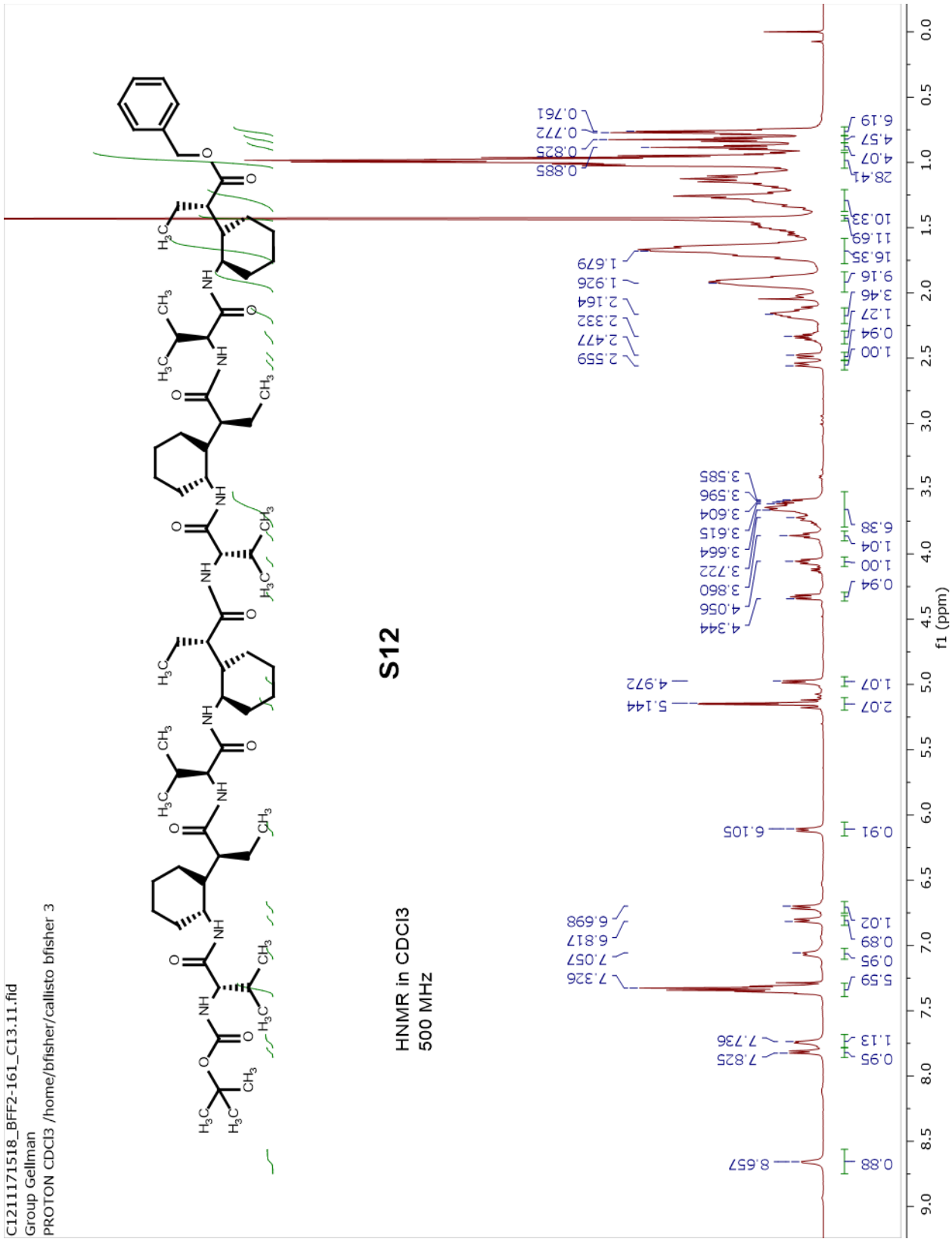

S10
HNMR in CDCl3
300 MHz
f1 (ppm)
0.000 TMS
7.925
7.691
7.477
7.266
0.85
0.86
3.03
2.11
0.80
6.880
0.83
6.002
5.167
5.068
2.02
1.00
1.04
4.577
0.98
3.846
3.27
3.682
2.295
2.184
4.33
1.905
1.678
7.61
7.47
10.34
26.30
3.74
0.991
0.913
0.757

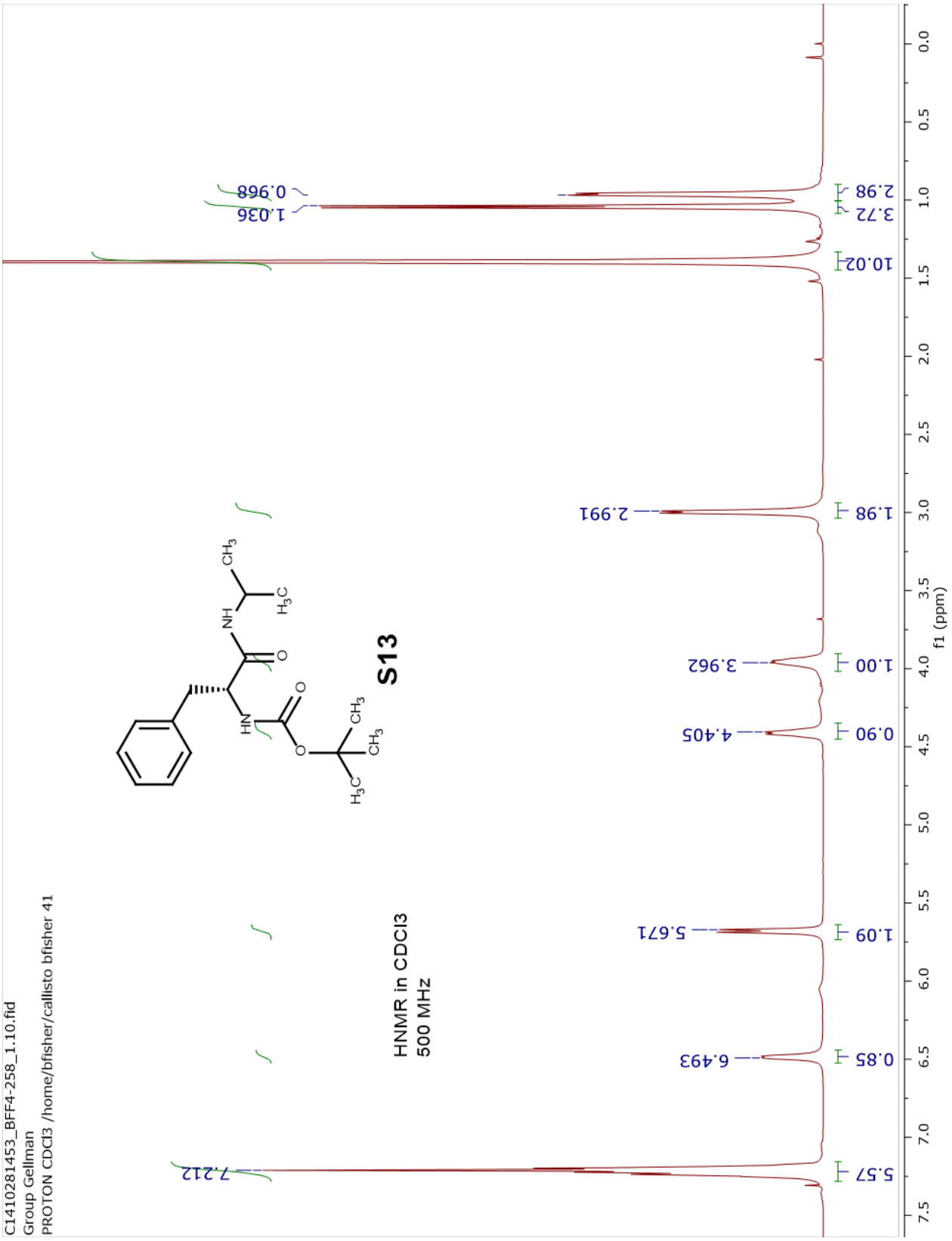

C130315107_BFF2-284_5mM.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 15
4
HNMR in CDCl3
500 MHz
f1 (ppm)

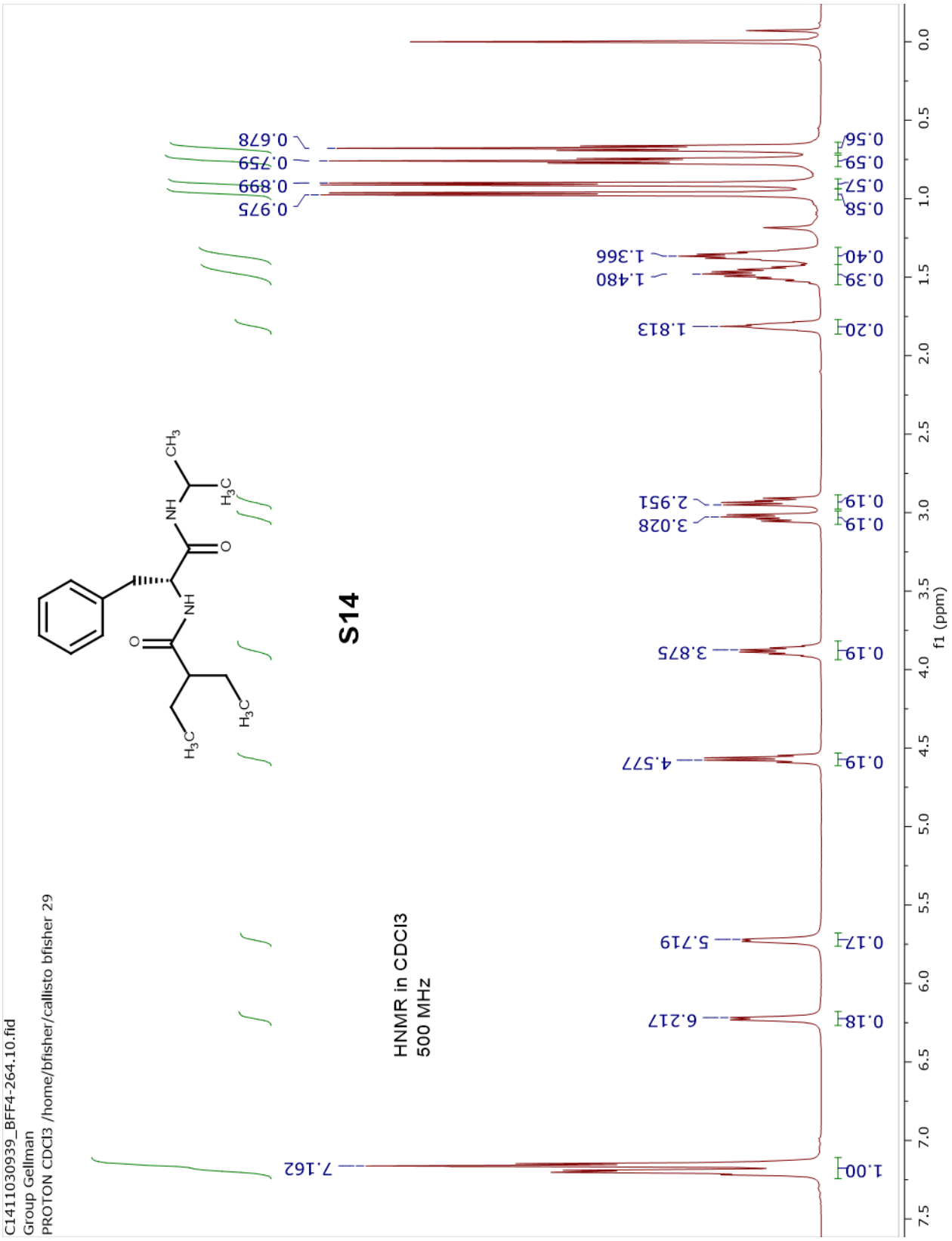

C1503091649_BFF4-082.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/tallisto bfisher 42
rac-5
HNMR in CDCl3
500 MHz
f1 (ppm)

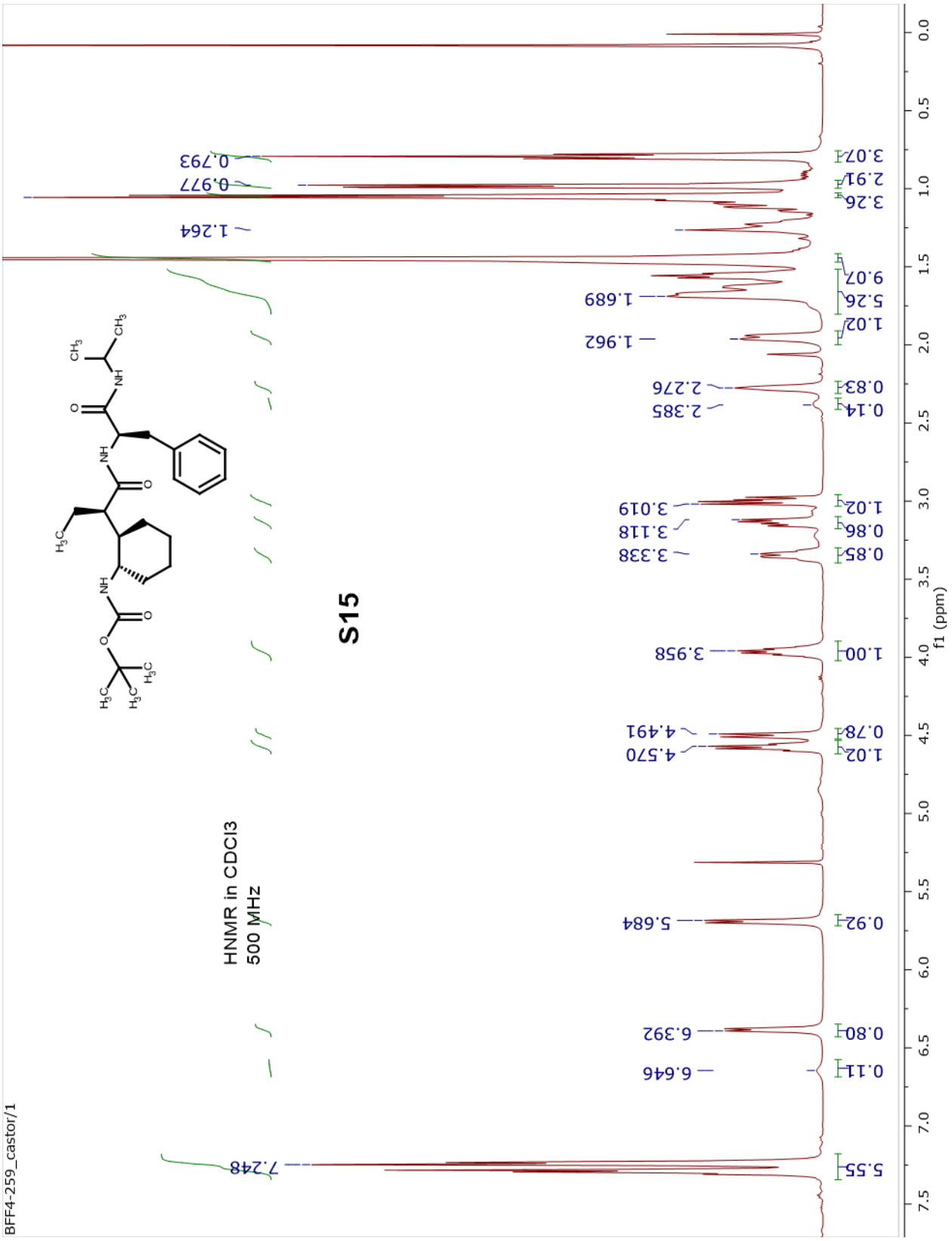

C1410241401_BFF4-253.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 19
HNMR in CDCl3
500 MHz
6
7.227
5.078
6.198
5.386
4.585
3.654
3.100
2.945
2.306
1.859
1.552
1.220
1.068
1.007
10.446
0.975
2.038
0.963
1.000
1.008
0.987
0.988
1.001
1.017
11.441
1.174
2.138
9.798
f1 (ppm)


S11
HNMR in CDCl3
400 MHz
D1301251126_BFF2-239-10mM.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/av400 bfisher 22
0.724
0.816
0.894
1.123
1.255
2.335
2.436
2.550
3.614
3.721
3.988
4.317
4.923
5.153
6.079
6.682
6.835
7.342
7.825
8.013
f1 (ppm)

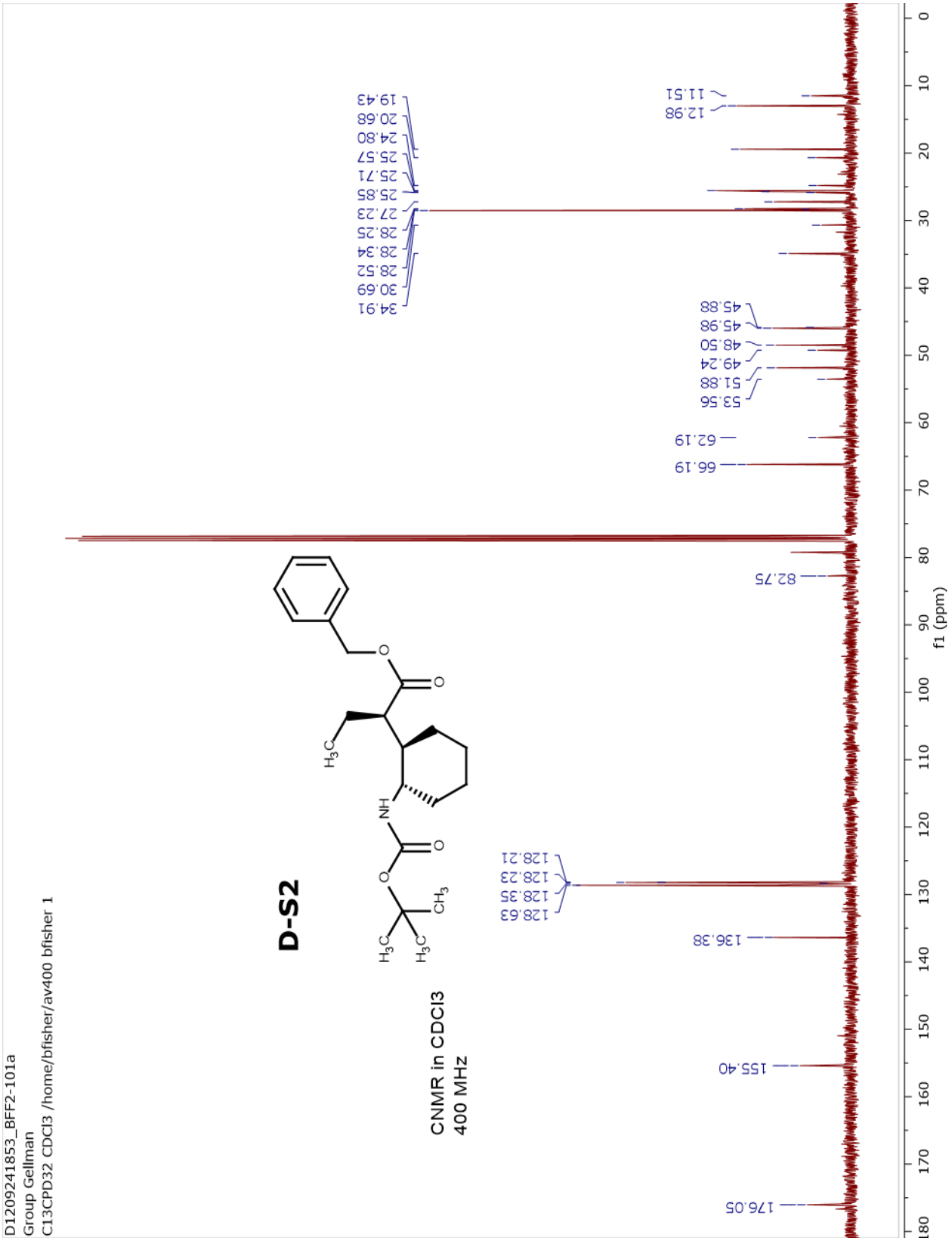

C1211171518_BFF2-161_C13.11.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 3
S12
HNMR in CDCl3
500 MHz
f1 (ppm)

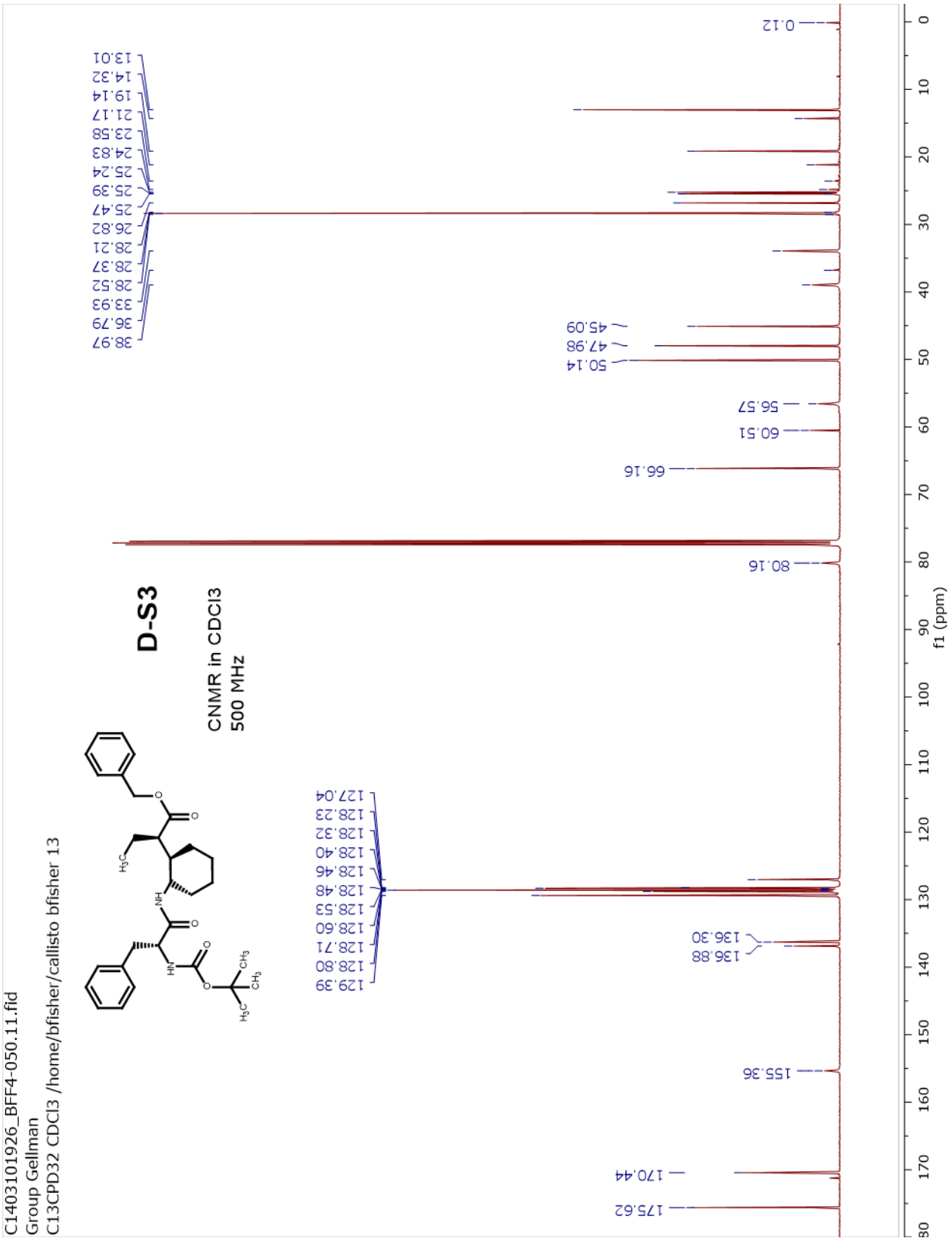

C1410281453_BFF4-258_1.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 41
HNMR in CDCl3
500 MHz
S13
7.212
6.493
5.671
4.405
3.962
2.991
1.036
0.968
5.57
0.85
1.09
0.90
1.00
1.98
10.02
3.72
2.98
f1 (ppm)

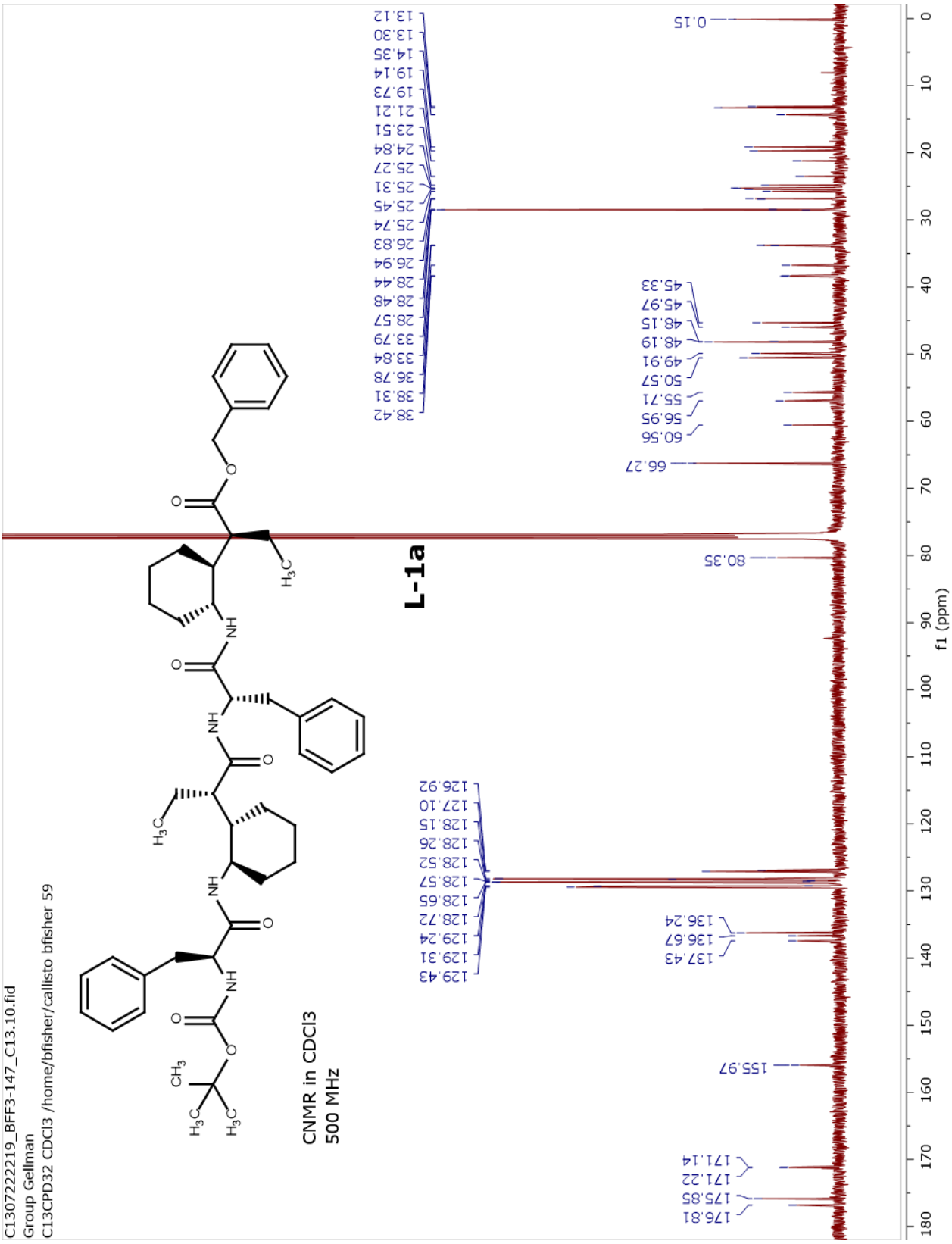

C1411030939_BFF4-264.10.fid
Group Gellman
PROTON CDCl3 /home/bfisher/callisto bfisher 29
HNMR in CDCl3
500 MHz
S14
7.162
6.217
5.719
4.577
3.875
3.028
2.951
1.813
1.480
1.366
0.975
0.899
0.759
0.678
1.00
0.18
0.17
0.19
0.19
0.19
0.19
0.20
0.39
0.40
0.58
0.57
0.59
0.56
f1 (ppm)

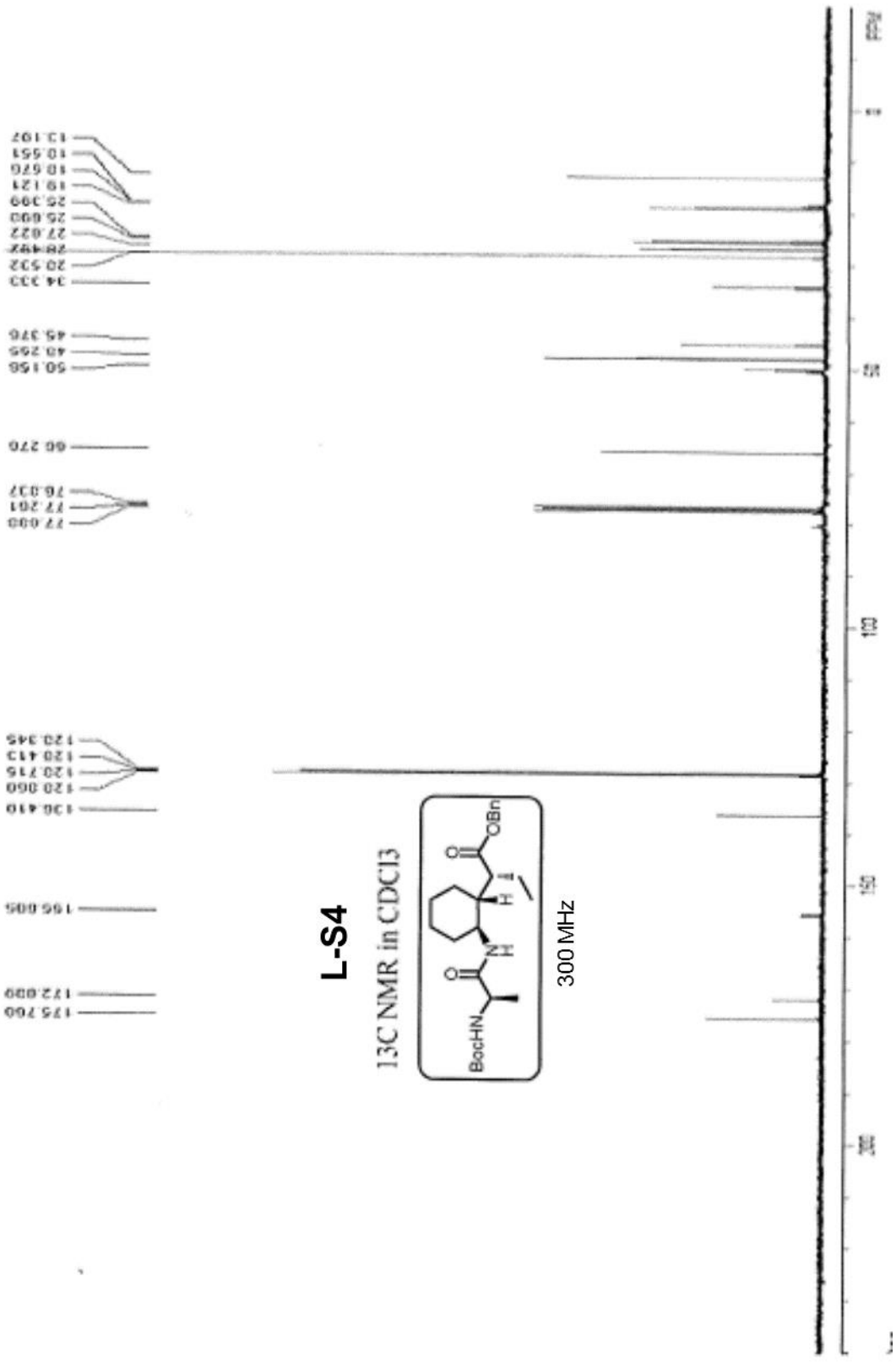

BFF4-259_castor/1
HNMR in CDCl3
500 MHz
S15
7.248
6.646
6.392
5.684
4.570
4.491
3.958
3.338
3.118
3.019
2.385
2.276
1.962
1.689
1.264
0.977
0.793
5.55
0.11
0.80
0.92
1.02
0.78
1.00
0.85
0.86
1.02
0.14
0.83
1.02
5.26
9.07
3.26
2.91
3.07
f1 (ppm)

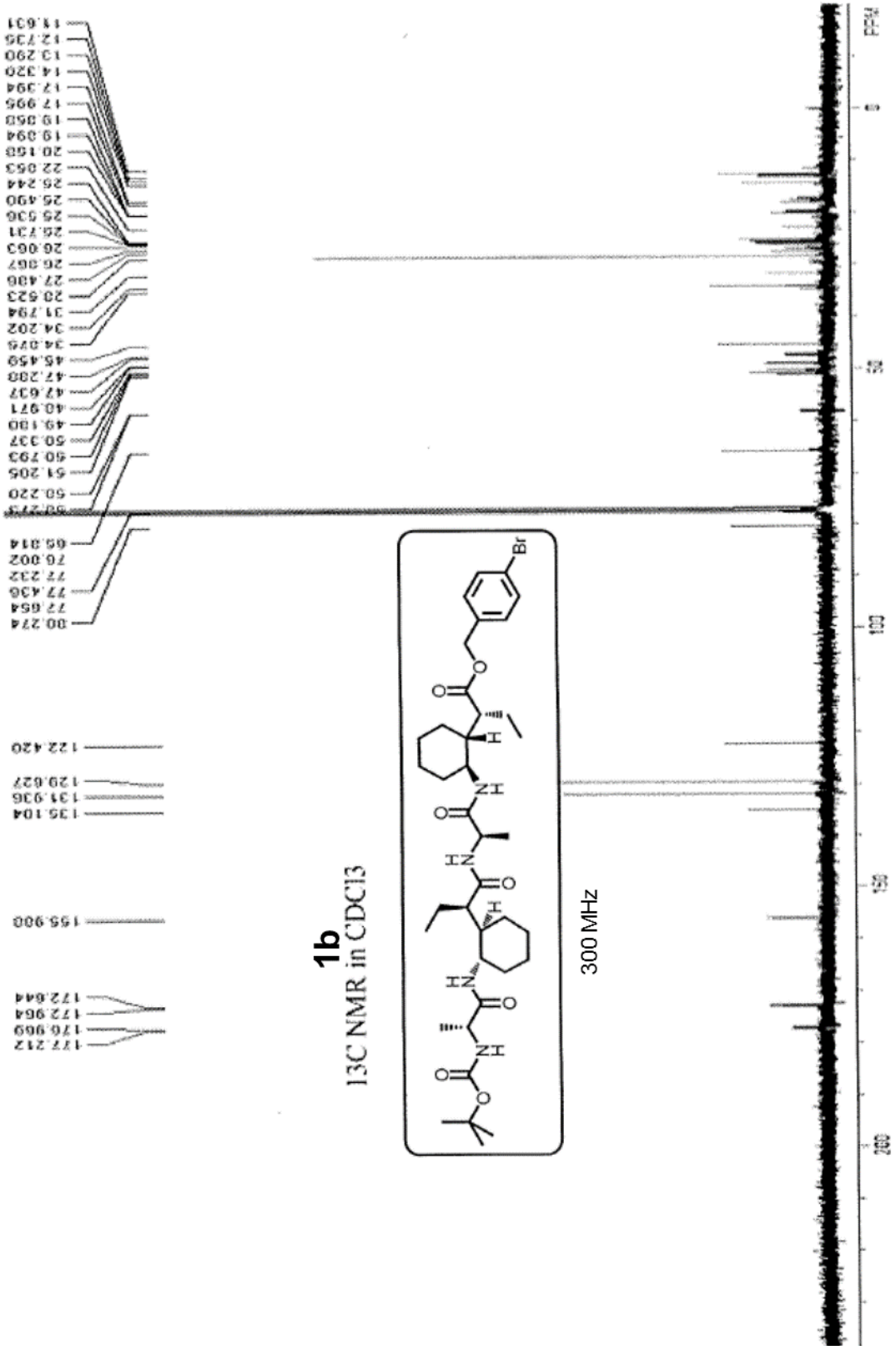
b. ^{13}C-NMR Spectra

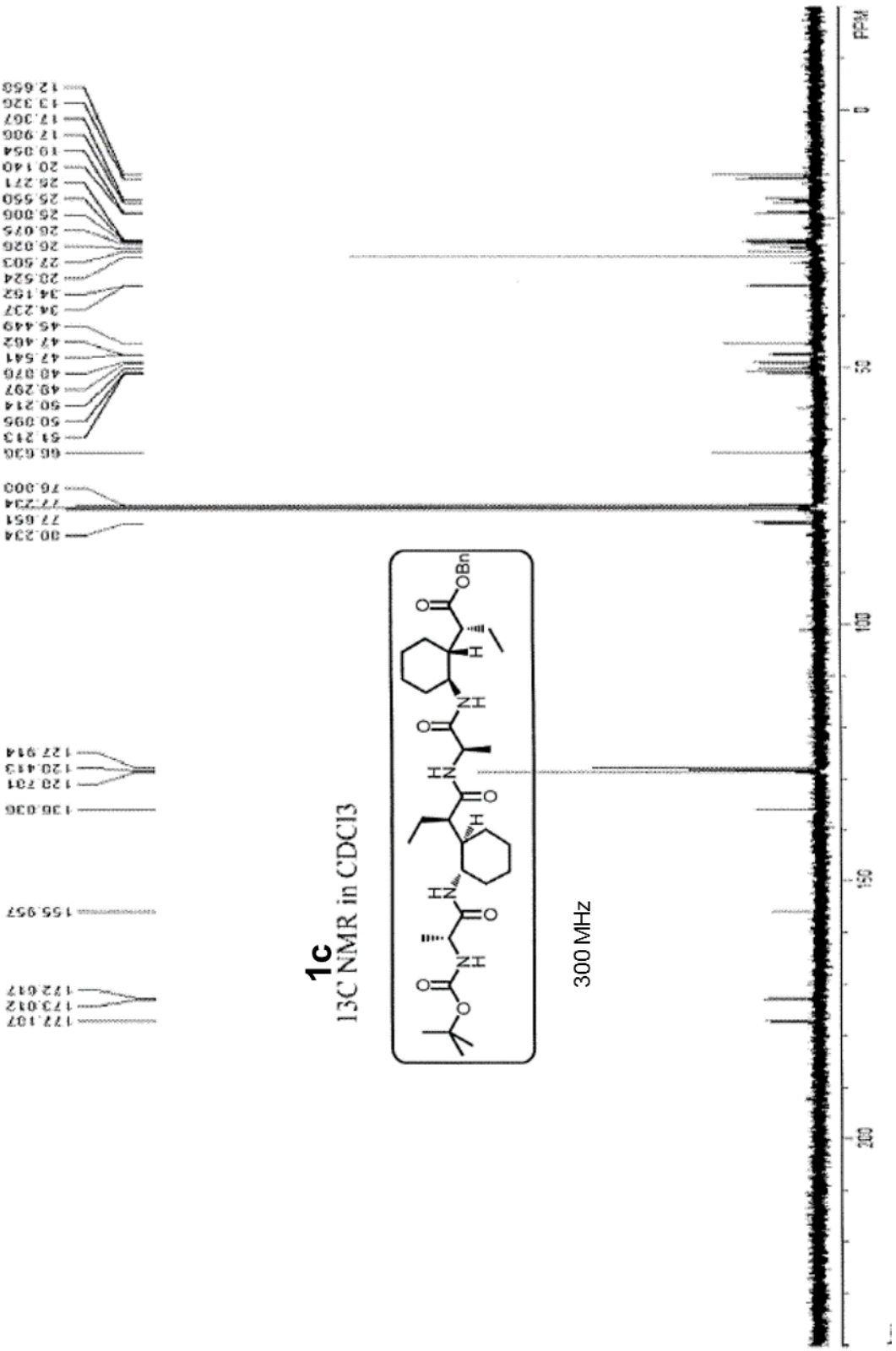


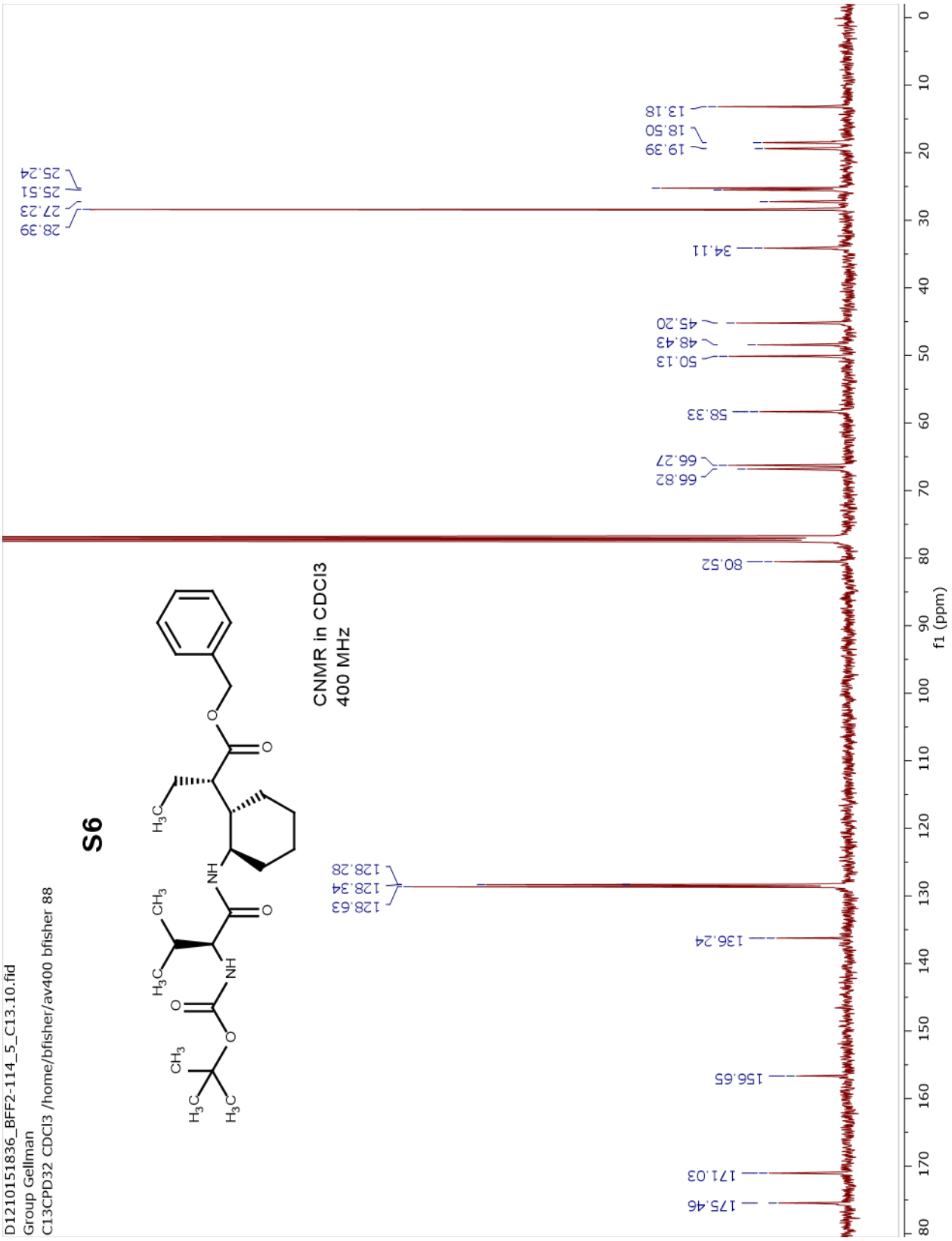

C140310926_BFF4-050.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 13
D-S3
CNMR in CDCl3
500 MHz
f1 (ppm)

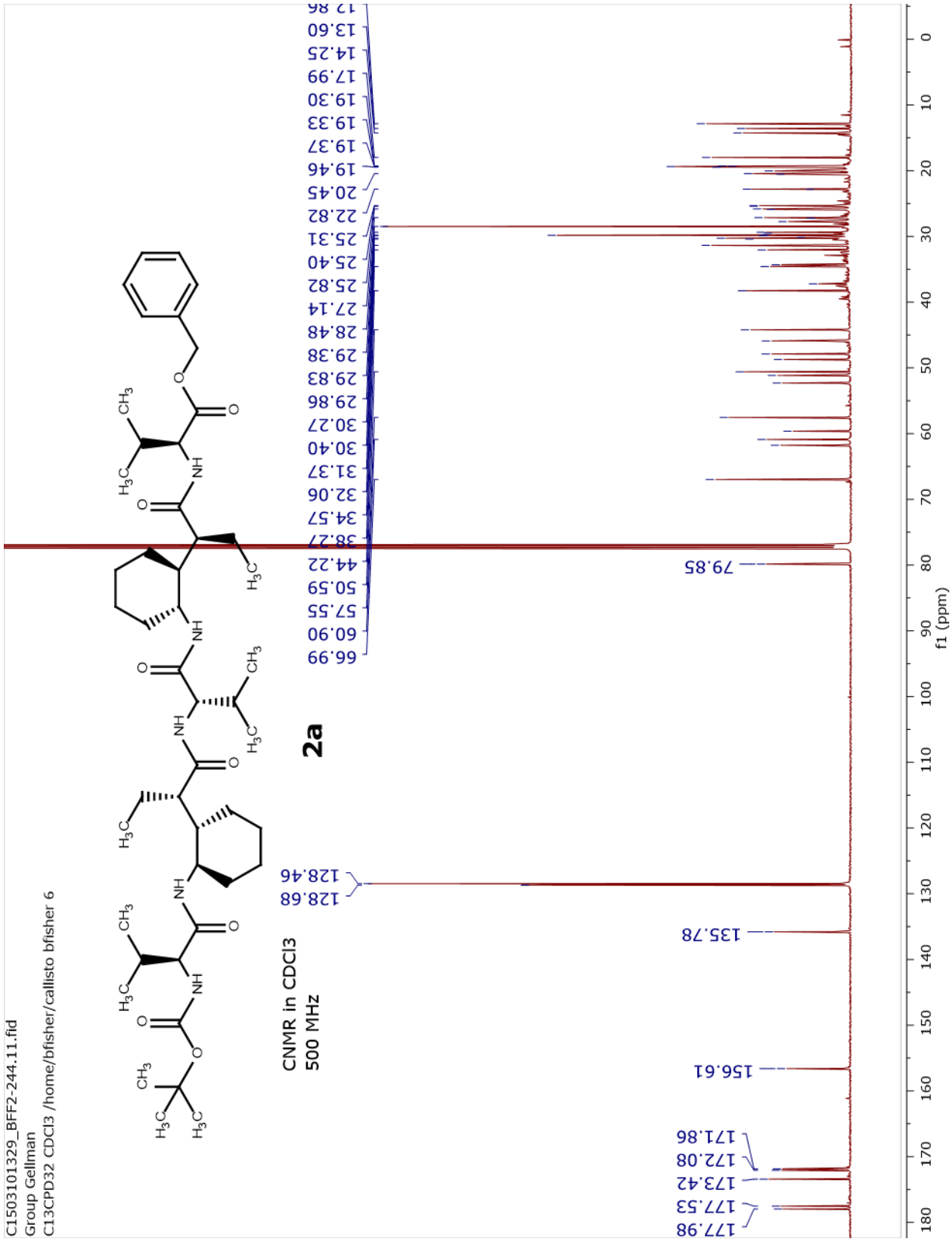

C1307222219_BFF3-147_C13.10.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 59
CNMR in CDCl3
500 MHz
L-1a
f1 (ppm)

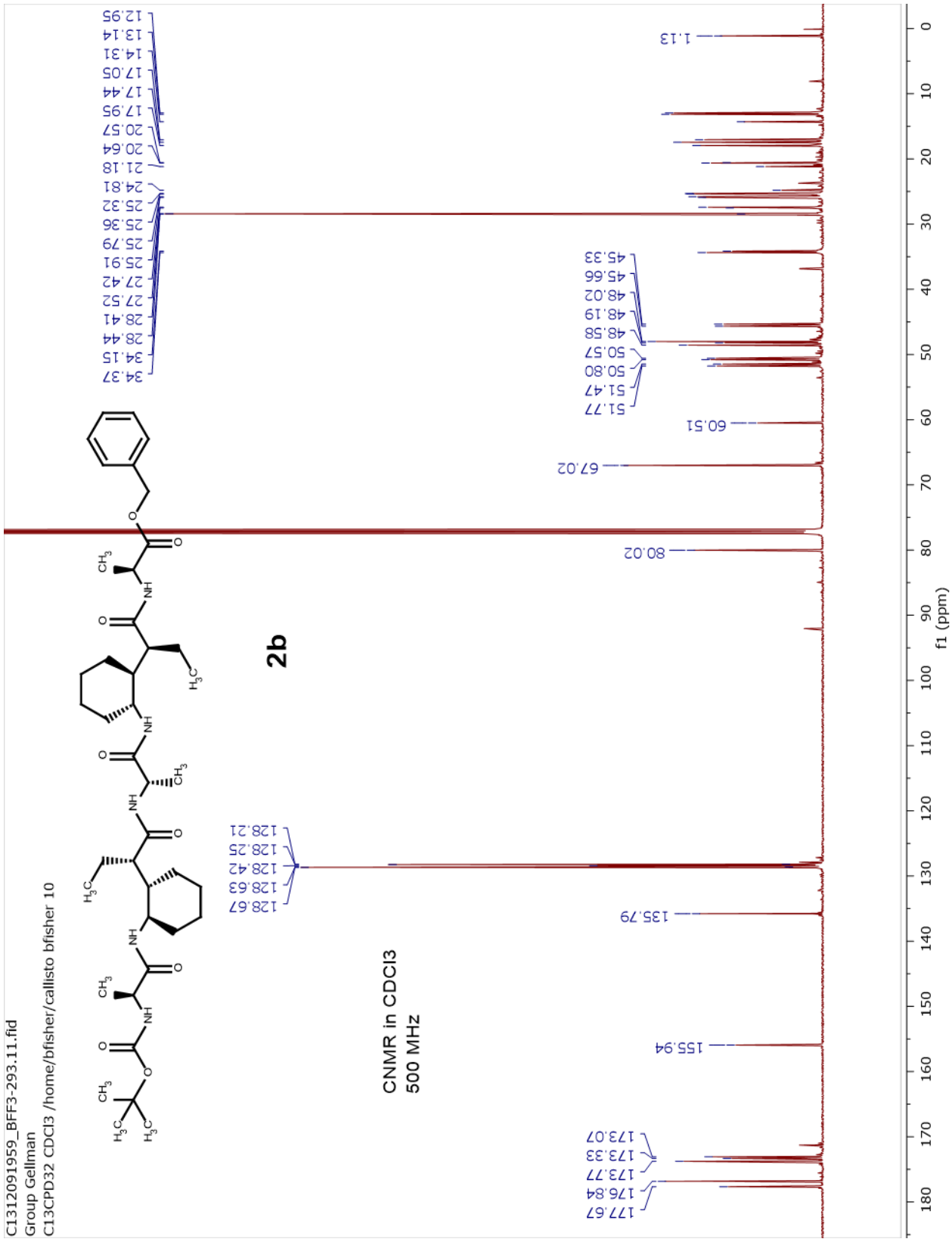

L-S4
13C NMR in CDCl3
300 MHz
BocHN
OBn
13.107
18.570
19.121
25.399
25.880
27.022
28.492
20.532
34.333
45.376
40.295
50.156
60.270
76.037
77.201
77.000
120.345
120.413
120.715
120.060
130.410
165.005
172.089
175.760

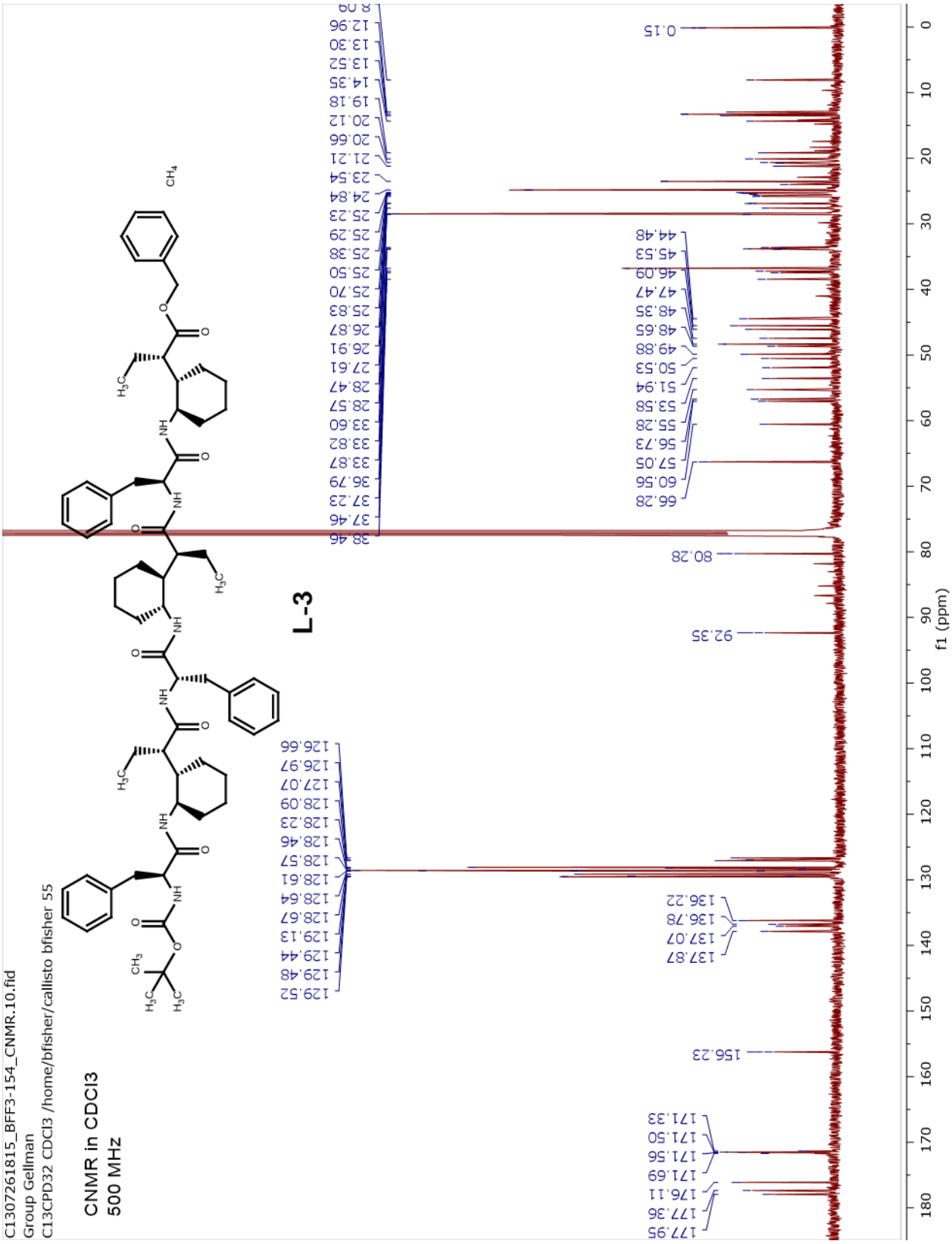

1b
13C NMR in CDCl3
300 MHz
Br
PPM

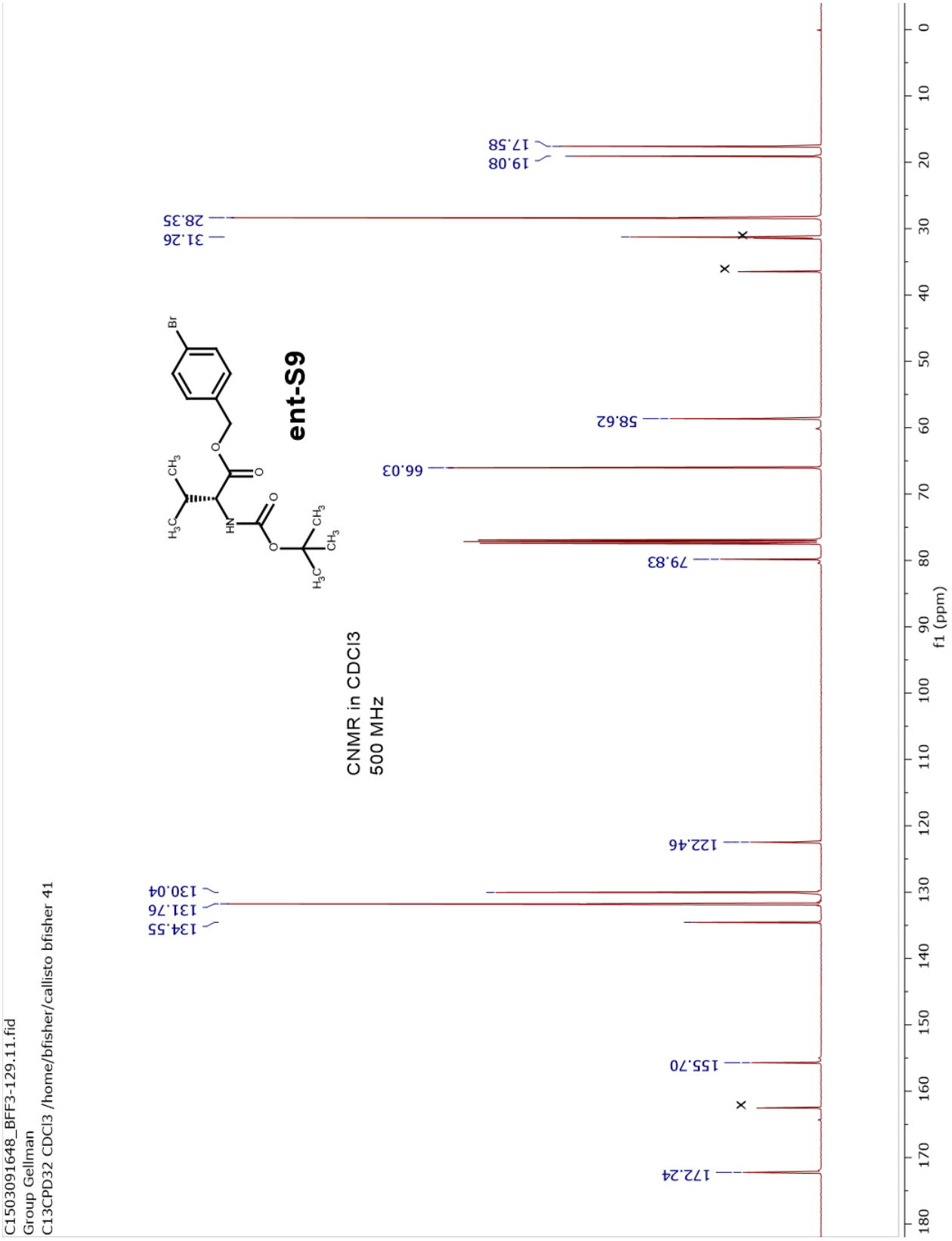

1c
13C NMR in CDCl3
300 MHz
OBn
PPM
0
50
100
150
200
12.650
13.326
17.367
17.906
19.054
20.140
26.271
25.550
25.806
26.075
26.826
27.503
20.524
34.152
34.237
45.449
47.462
47.541
48.076
48.297
50.214
50.095
51.213
66.636
76.000
77.234
77.651
80.234
127.914
128.413
128.781
136.036
155.957
172.617
173.012
177.107

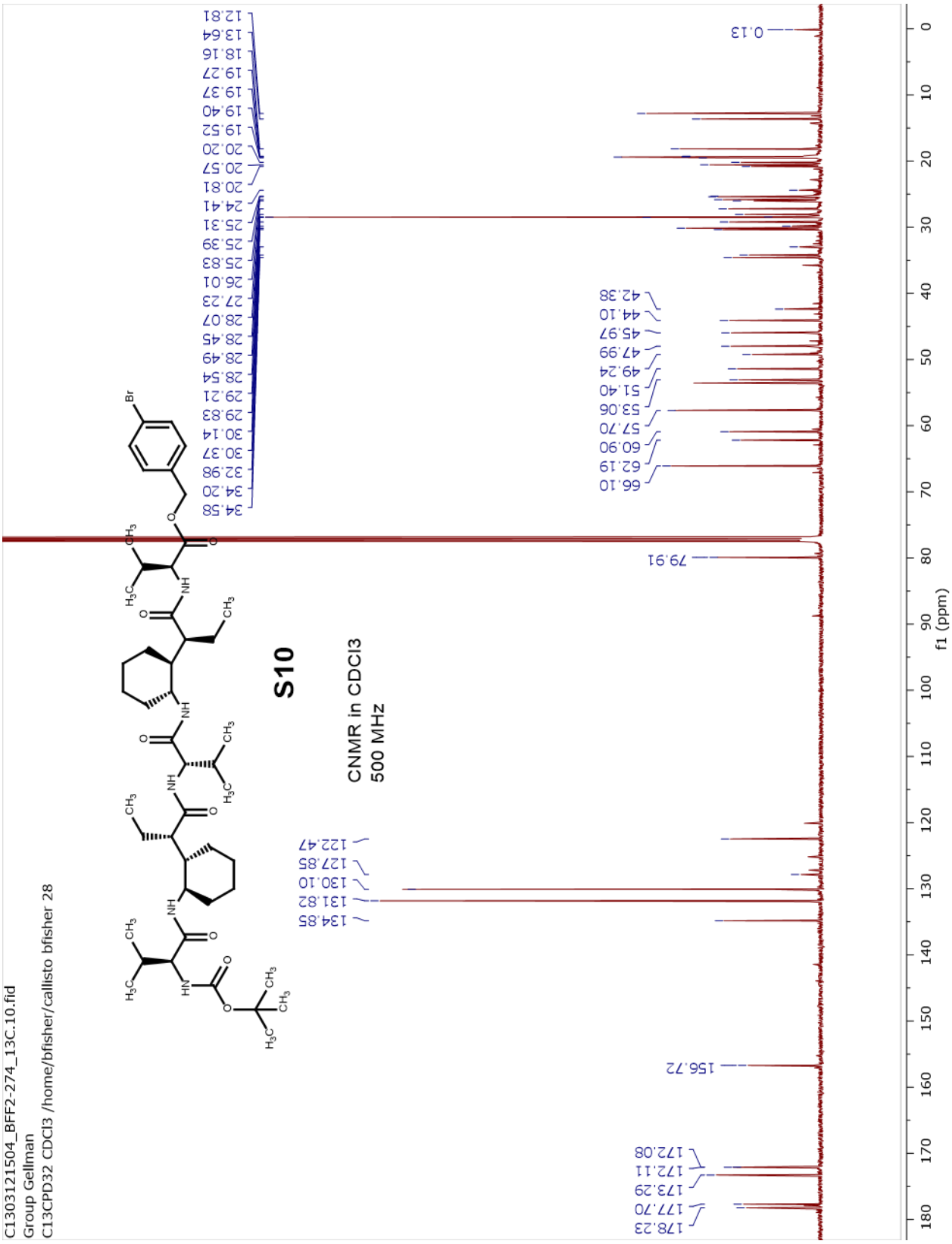

D1210151836_BFF2-114_5_C13.10.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/av400 bfisher 88
S6
CNMR in CDCl3
400 MHz
175.46
171.03
156.65
136.24
128.63
128.34
128.28
80.52
66.82
66.27
58.33
50.13
48.43
45.20
34.11
28.39
27.23
25.51
25.24
19.39
18.50
13.18
f1 (ppm)


C150310329_BFF2-244.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 6
CNMR in CDCl3
500 MHz
2a
f1 (ppm)


C131209195 9_BFF3-293.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 10
2b
CNMR in CDCl3
500 MHz
f1 (ppm)


C130726181S_BFF3-154_CNMR.10.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 55
CNMR in CDCl3
500 MHz
L-3
f1 (ppm)


C150309l648_BFF3-129.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 41
CNMR in CDCl3
500 MHz
ent-S9
Br
CH3
H3C
O
HN
O
O
CH3
CH3
H3C
172.24
155.70
134.55
131.76
130.04
122.46
79.83
66.03
58.62
31.26
28.35
19.08
17.58
f1 (ppm)


C130312504_BFF2-274_13C.10.fid
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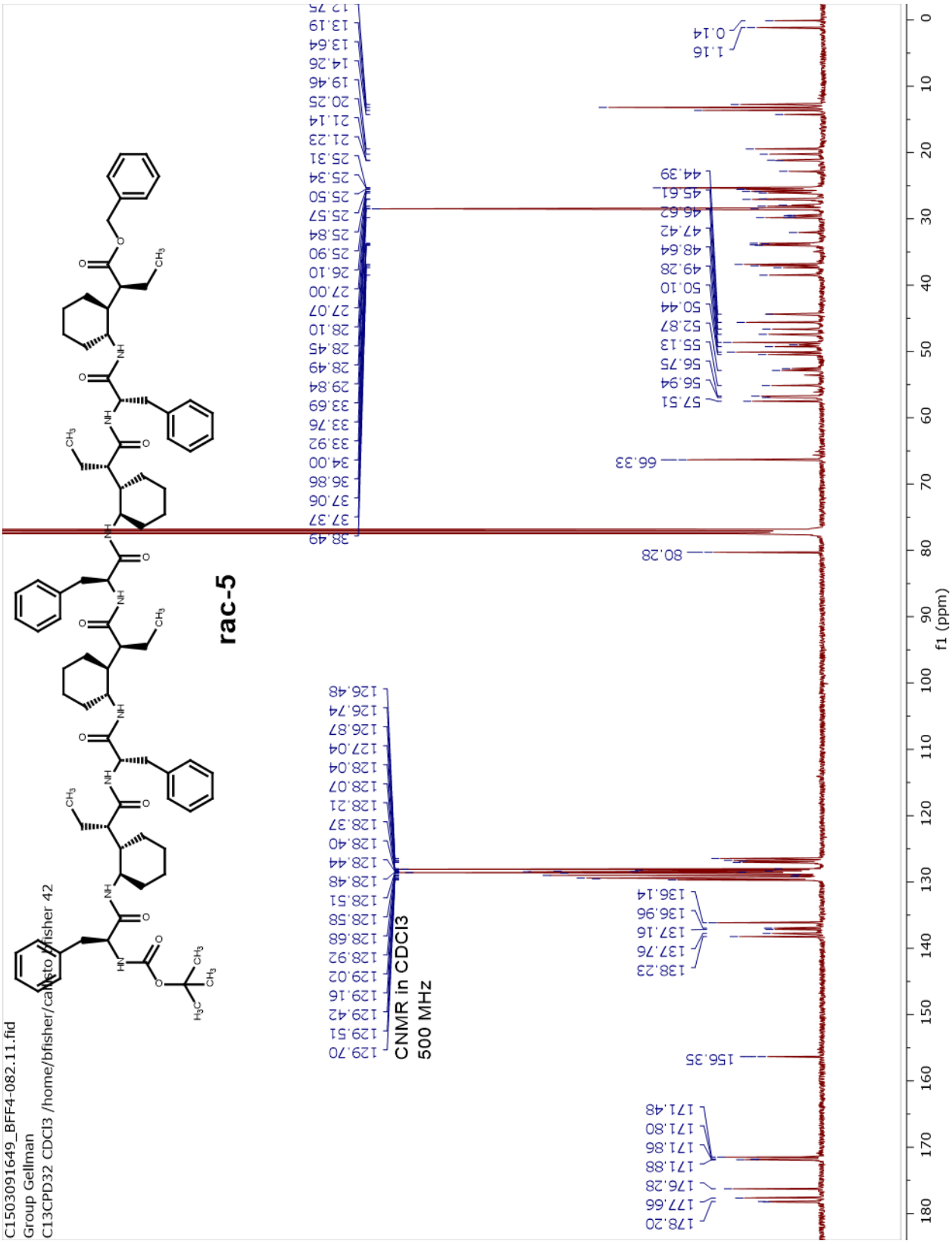
S10
CNMR in CDCl3
500 MHz
f1 (ppm)


C1503091649_BFF4-082.11.fid
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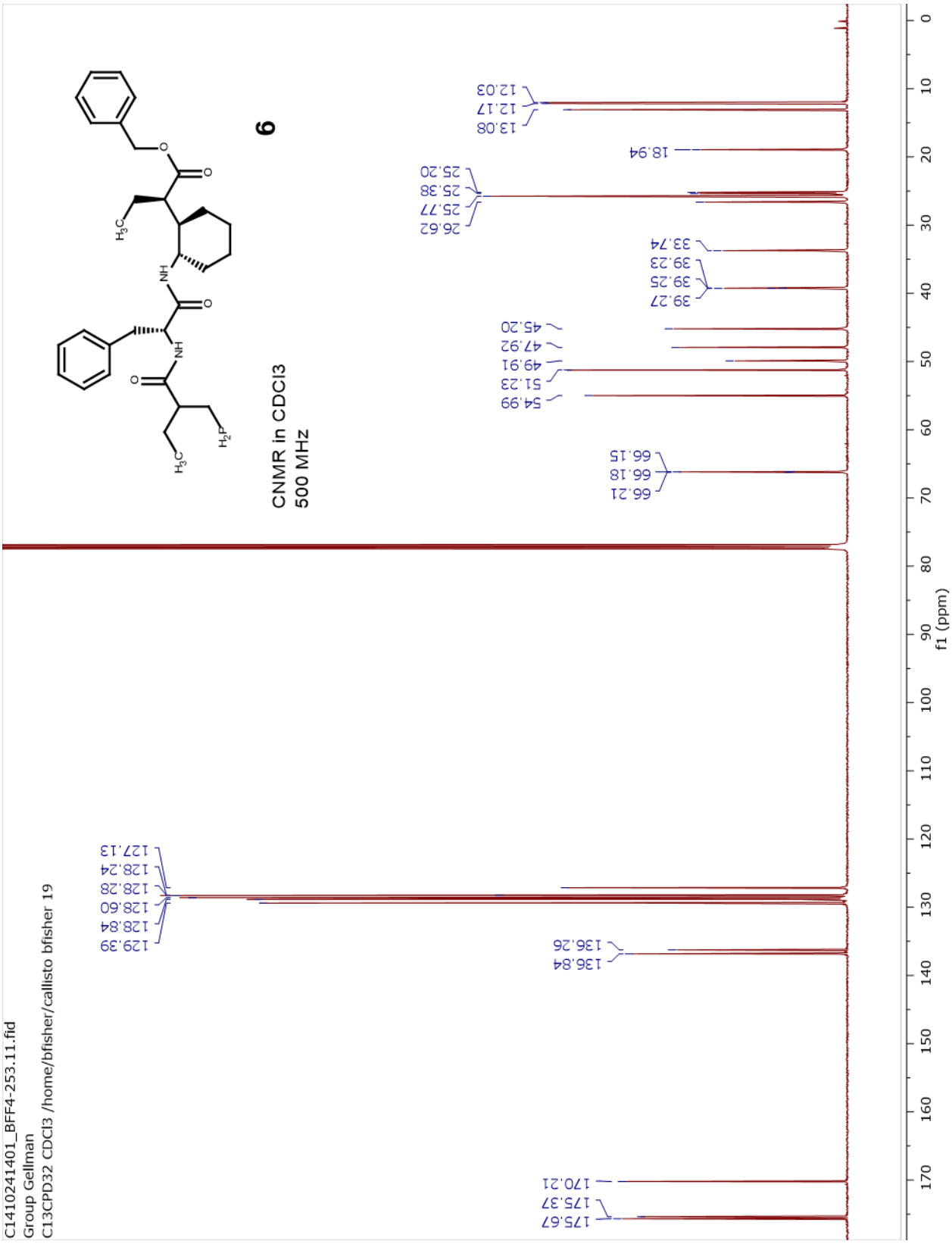
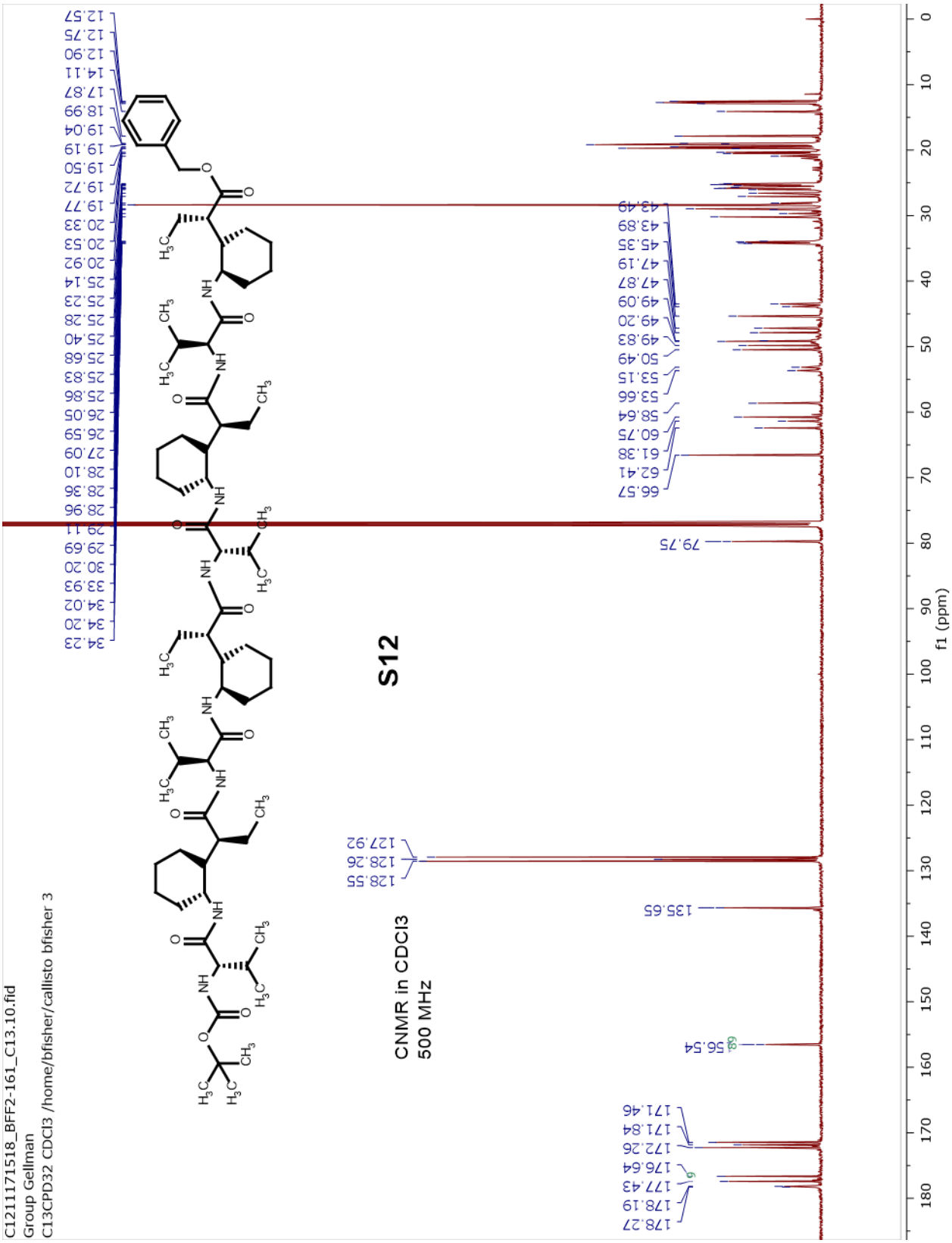
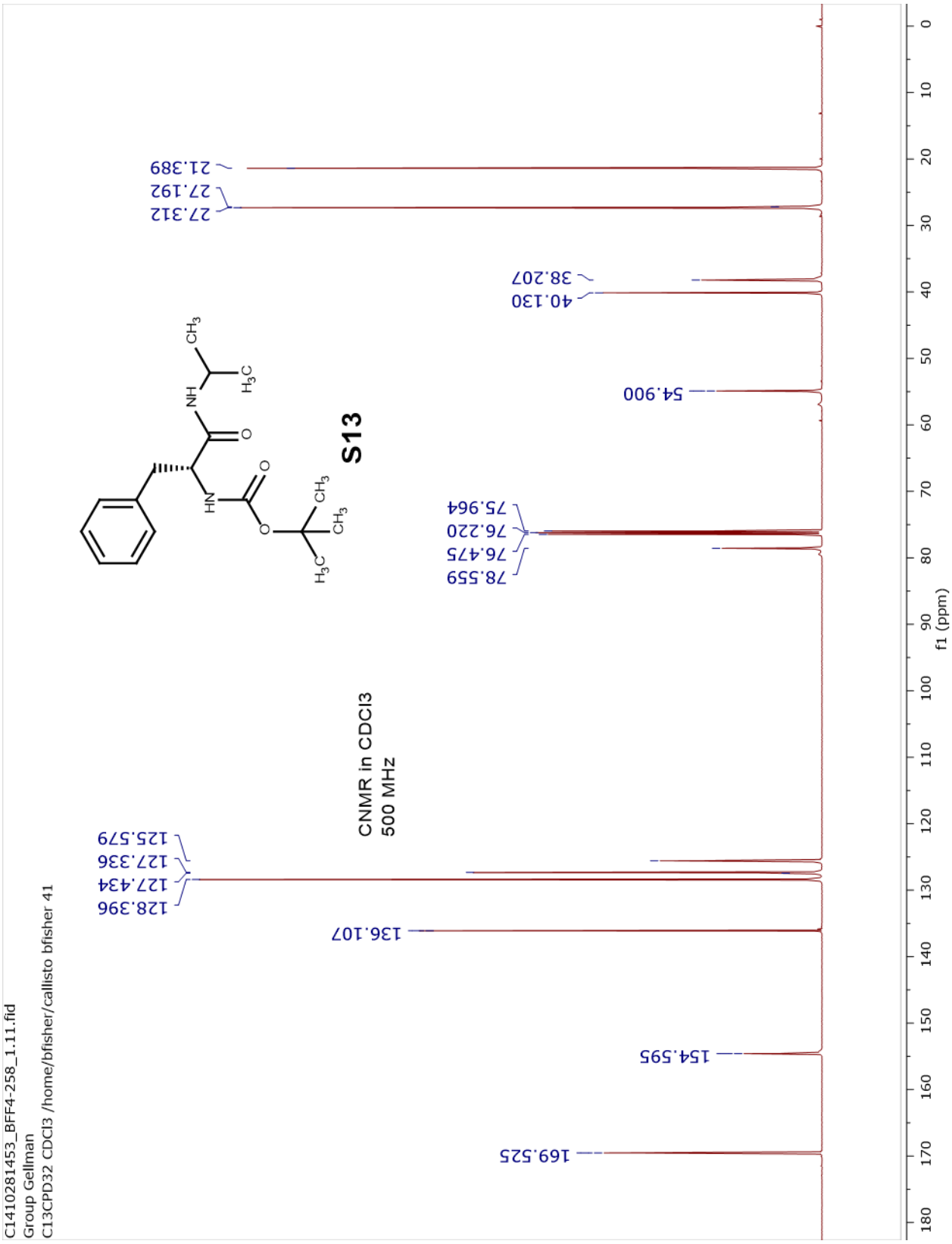
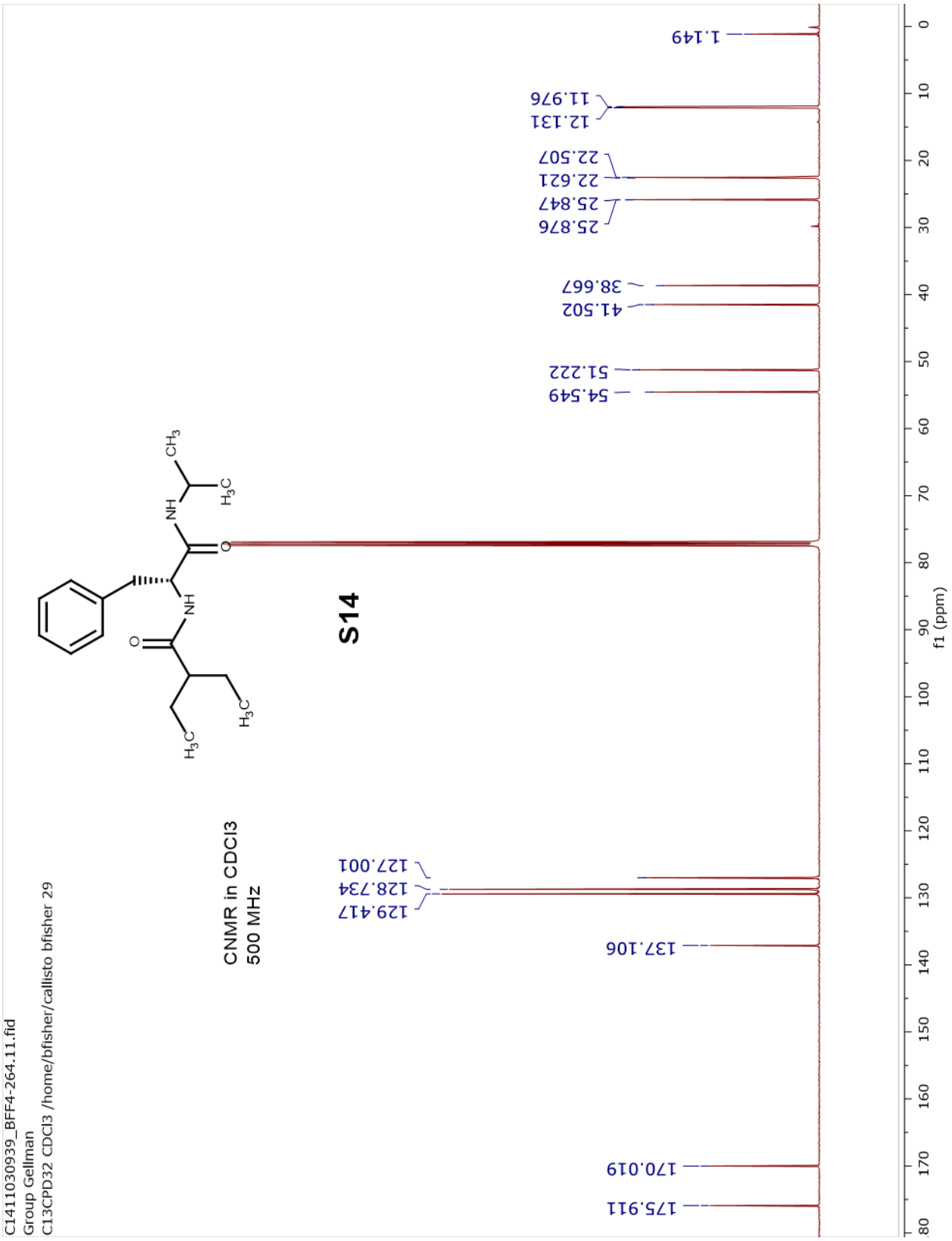
rac-5
CNMR in CDCl3
500 MHz
f1 (ppm)


C1410241401_BFF4-253.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 19
6
CNMR in CDCl3
500 MHz
175.67
175.37
170.21
136.84
136.26
129.39
128.84
128.60
128.28
128.24
127.13
66.21
66.18
66.15
54.99
51.23
49.91
47.92
45.20
39.27
39.25
39.23
33.74
26.62
25.77
25.38
25.20
18.94
13.08
12.17
12.03
f1 (ppm)


C1211171518_BFF2-161_C13.10.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 3
S12
CNMR in CDCl3
500 MHz
f1 (ppm)


C1410281453_BFF4-258_1.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 41
CNMR in CDCl3
500 MHz
S13
21.389
27.192
27.312
38.207
40.130
54.900
75.964
76.220
76.475
78.559
125.579
127.336
127.434
128.396
136.107
154.595
169.525
f1 (ppm)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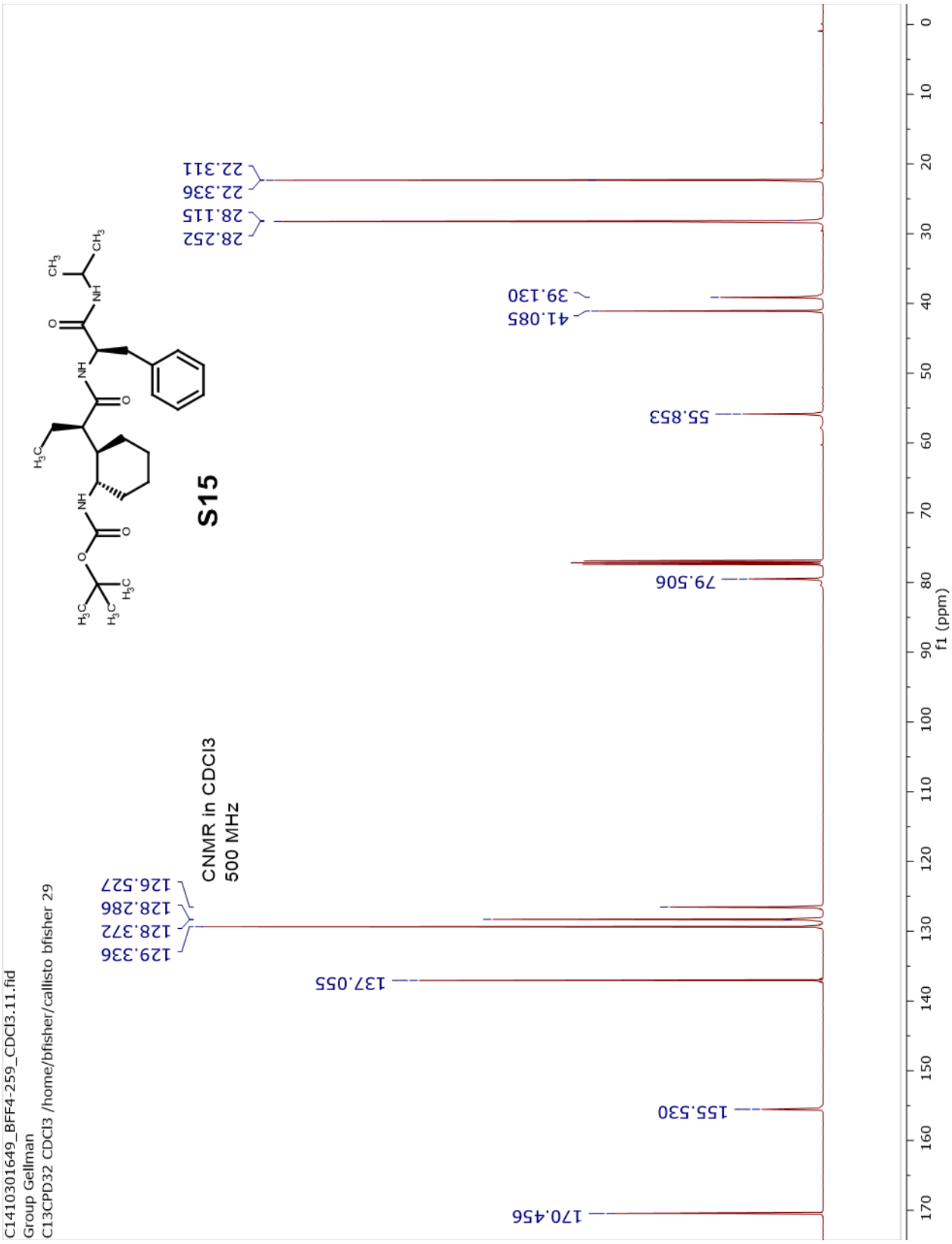
CNMR in CDCl3
500 MHz
S14
1.149
11.976
12.131
22.507
22.621
25.847
25.876
38.667
41.502
51.222
54.549
127.001
128.734
129.417
137.106
170.019
175.911
f1 (ppm)


C141030164 9_BFF4-259_CDCl3.11.fid
Group Gellman
C13CPD32 CDCl3 /home/bfisher/callisto bfisher 29
CNMR in CDCl3
500 MHz
S15
22.311
22.336
28.115
28.252
39.130
41.085
55.853
79.506
126.527
128.286
128.372
129.336
137.055
155.530
170.456
f1 (ppm)

c. 2D-NMR Spectra

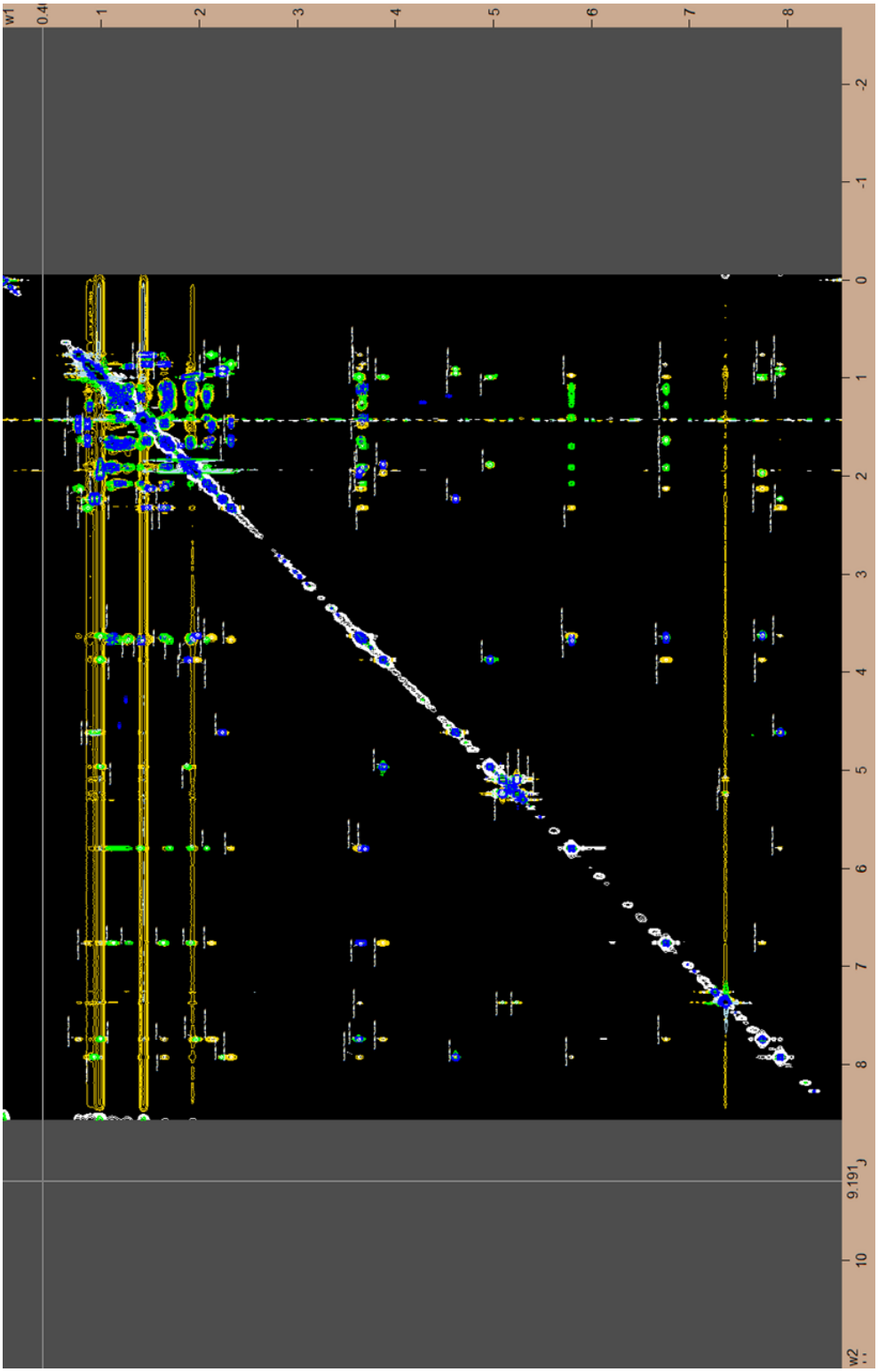


Figure S14. Overlay of 2D-NMR Spectra of **2a**. Blue is COSY, green is TOCSY, and gold/white is NOESY positive/negative.

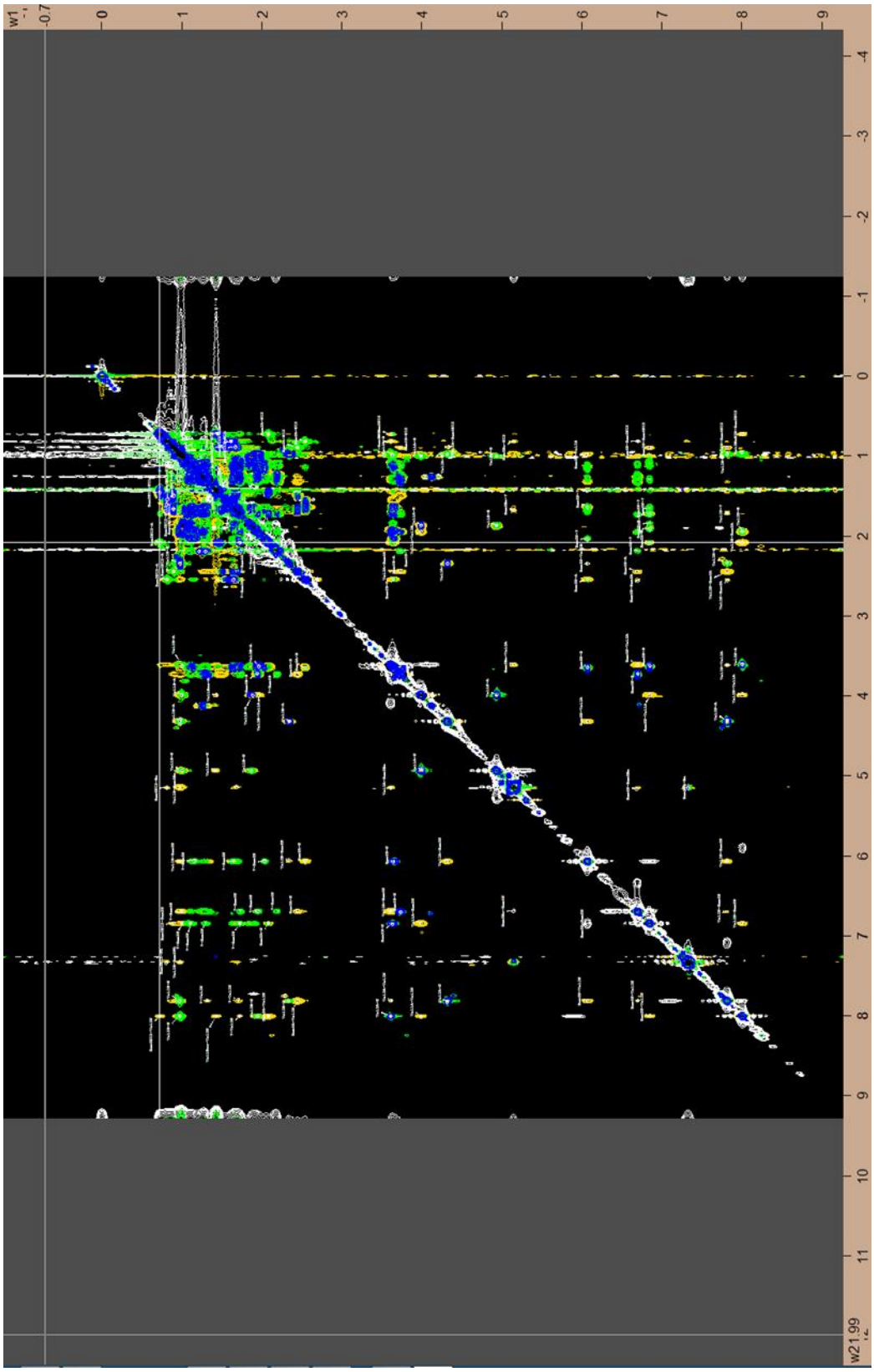


Figure S15. Overlay of 2D-NMR Spectra of **S11**. Blue is COSY, green is TOCSY, and gold/white is NOESY positive/negative.

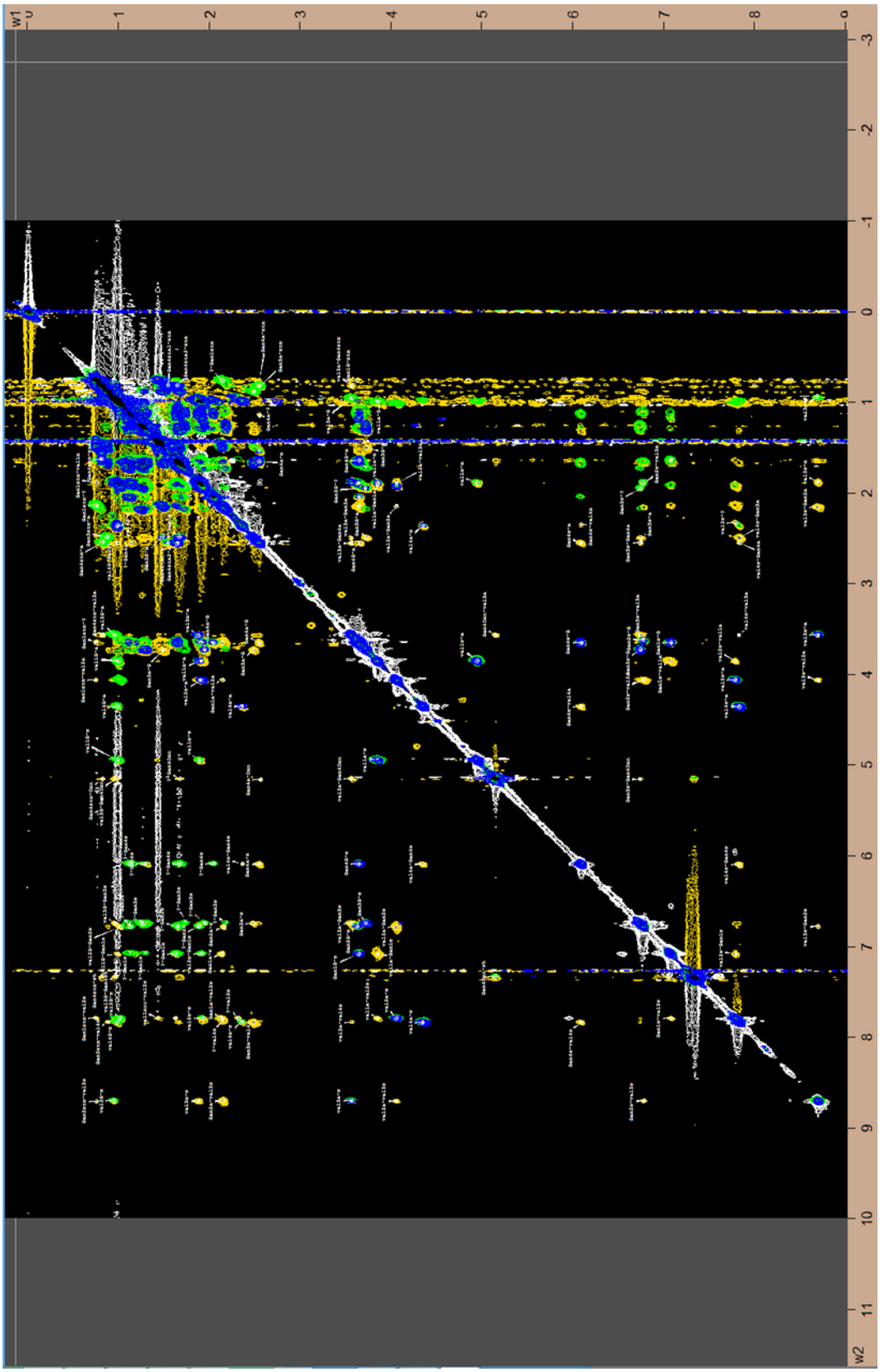


Figure S16. Overlay of 2D-NMR Spectra of **S12**. Blue is COSY, green is TOCSY, and gold/white is ROESY positive/negative.

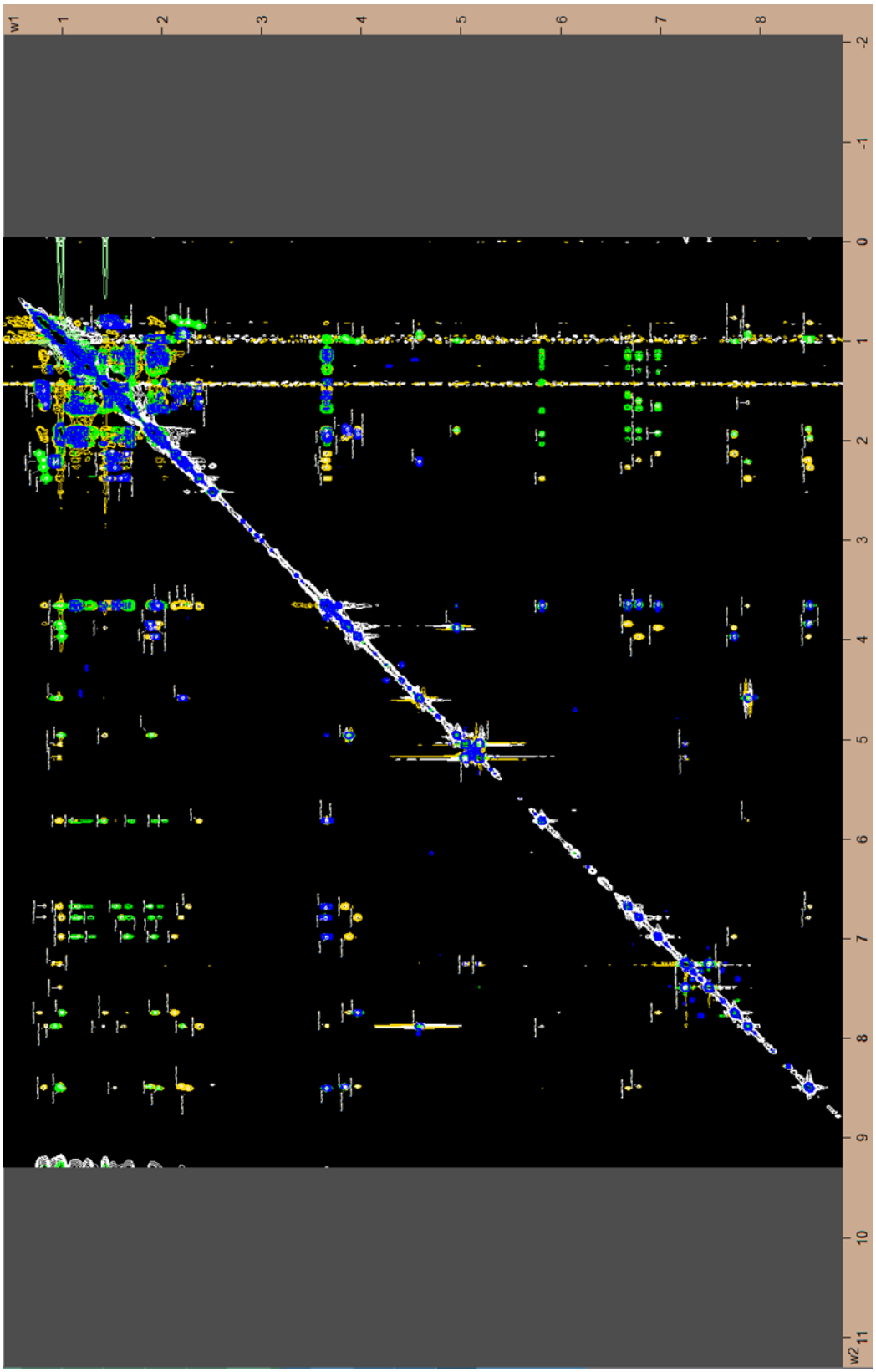


Figure S17. Overlay of 2D-NMR Spectra of **4**. Blue is COSY, green is TOCSY, and gold/white is ROESY positive/negative.

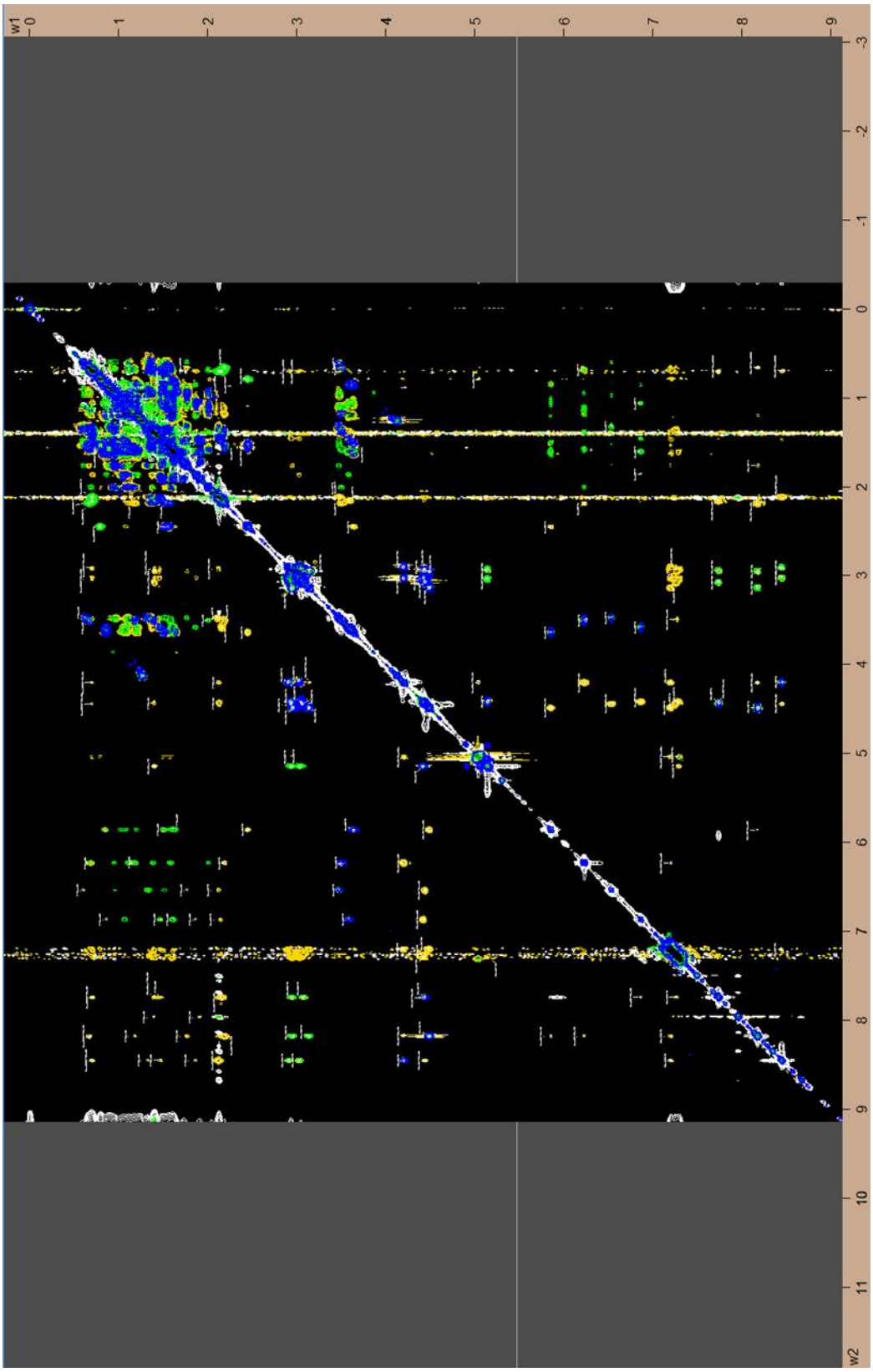


Figure S18. Overlay of 2D-NMR Spectra of **5**. Blue is COSY, green is TOCSY, and gold/white is ROESY positive/negative.

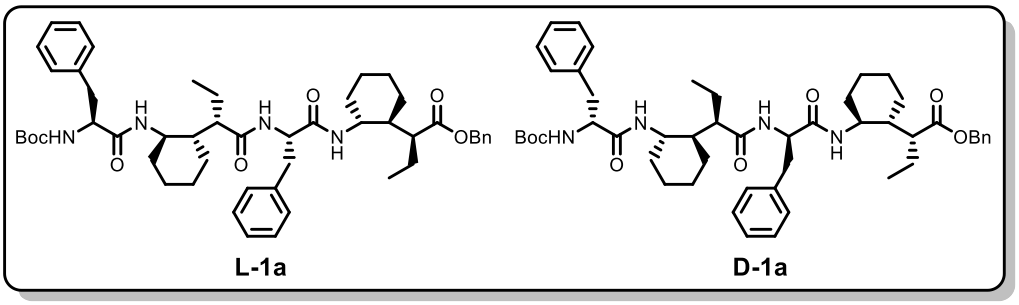
XI. Crystallographic Data

Crystals grown by the slow evaporation method were done in loosely-capped 2 mL scintillation vials in solutions of 150 μL to 1 mL solvent. Crystals grown by the vapor diffusion method were done by dissolving peptide in the strongly-solvating solvent in a 2 mL scintillation vial and placing this vial into a 15 mL scintillation vial containing volatile precipitating solvent. This larger vial was tightly capped. Crystals grown at 4°C were grown inside a chemical refrigerator. Racemic crystallization was done by transferring approximately equal amounts of each peptide enantiomer into a 2 mL scintillation vial. Each crystallized racemate formed a centrosymmetric space group.

Table S19. Crystallization setups

Crystal	Method	Solvent system	Temperature
1a	Racemic; slow evaporation	1:6 CHCl3:heptane	rt
1b	Slow evaporation	EtOAc:heptane:MeOH	rt
1c	Slow evaporation	Et2O:heptane	rt
2a	Slow evaporation	1:8 CHCl3:heptane	4°C
2b	Slow evaporation	1:3 EtOH:heptane	rt
3a	Racemic; vapor diffusion	pentane into 3:1 CHCl3:DCE	4°C
3b	Racemic; vapor diffusion	pentane into CHCl3	4°C

Crystallographic Experimental Section for Compound *rac*-1a



Data Collection

A colorless crystal with approximate dimensions 0.162 × 0.119 × 0.091 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker Quazar SMART APEXII diffractometer with Mo K_α ($\lambda = 0.71073$ Å) radiation and the diffractometer to crystal distance of 4.96 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 12 frames collected at intervals of 0.5º in a 6º range about ω with the exposure time of 20 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program suite. The final cell constants were calculated from a set of 9493 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.70 Å. A total of 22130 data were harvested by collecting 5 sets of frames with 0.5º scans in ω and φ with exposure times of 90 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were consistent for the space groups $P\bar{1}$ and $P1$. The *E*-statistics strongly suggested the centrosymmetric space group $P\bar{1}$ that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms were refined with anisotropic displacement coefficients unless otherwise specified. All aromatic and aliphatic hydrogen atoms were included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

The structure is racemic.

The Ph ring at carbon C7 is disordered over two positions with the major component present 91.9(4)% of the time. The minor component was refined isotropically with an idealized geometry.

There is also 0.8161(16) molecule of solvate chloroform and 0.1839(16) molecules of heptane per foldamer in the crystals. The solvent molecules share the same site resulting in compositional disorder. The heptane was refined isotropically with constraints and restraints.

The final least-squares refinement of 640 parameters against 15641 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0546 and 0.1433, respectively. The final difference Fourier map was featureless.

Summary

Crystal Data for $C_{52.1}H_{71.8}Cl_{2.4}N_4O_7$ (M =952.90): triclinic, space group $P\bar{1}$ (no. 2), a = 11.5447(4) Å, b = 14.2478(5) Å, c = 17.4006(6) Å, α = 96.227(2)°,β = 108.4600(18)°, γ = 105.9040(18)°, V = 2550.47(16) Å^3, Z = 2, T = 100.0 K, μ(MoKα) = 0.204 mm^{-1}, *Dcalc* = 1.241 g/mm^3, 62220 reflections measured ($2.526 \leq 2\Theta \leq 61.23$), 15641 unique ($R_{int}$ = 0.0557, R_{sigma} = 0.0599) which were used in all calculations. The final R_1 was 0.0546 ($I > 2\sigma(I)$) and wR_2 was 0.1433 (all data).

References

[1] Bruker-AXS. (2007-2014) APEX2 (Ver. 2014.1-1), SADABS (2012-1), and SAINT+ (Ver. 8.32A) Software Reference Manuals. Bruker-AXS, Madison, Wisconsin, USA.

[2] Sheldrick, G. M. (2008) SHELXL. *Acta Cryst.* **A64**, 112-122.

[3] Dolomanov, O.V.; Bourhis, L.J.; Gildea, R.J.; Howard, J.A.K.; Puschmann, H. "OLEX2: a complete structure solution, refinement and analysis program". *J. Appl. Cryst.* (2009) **42**, *339-341*.

[4] Guzei, I.A. (2013). Internal laboratory computer programs Gn.

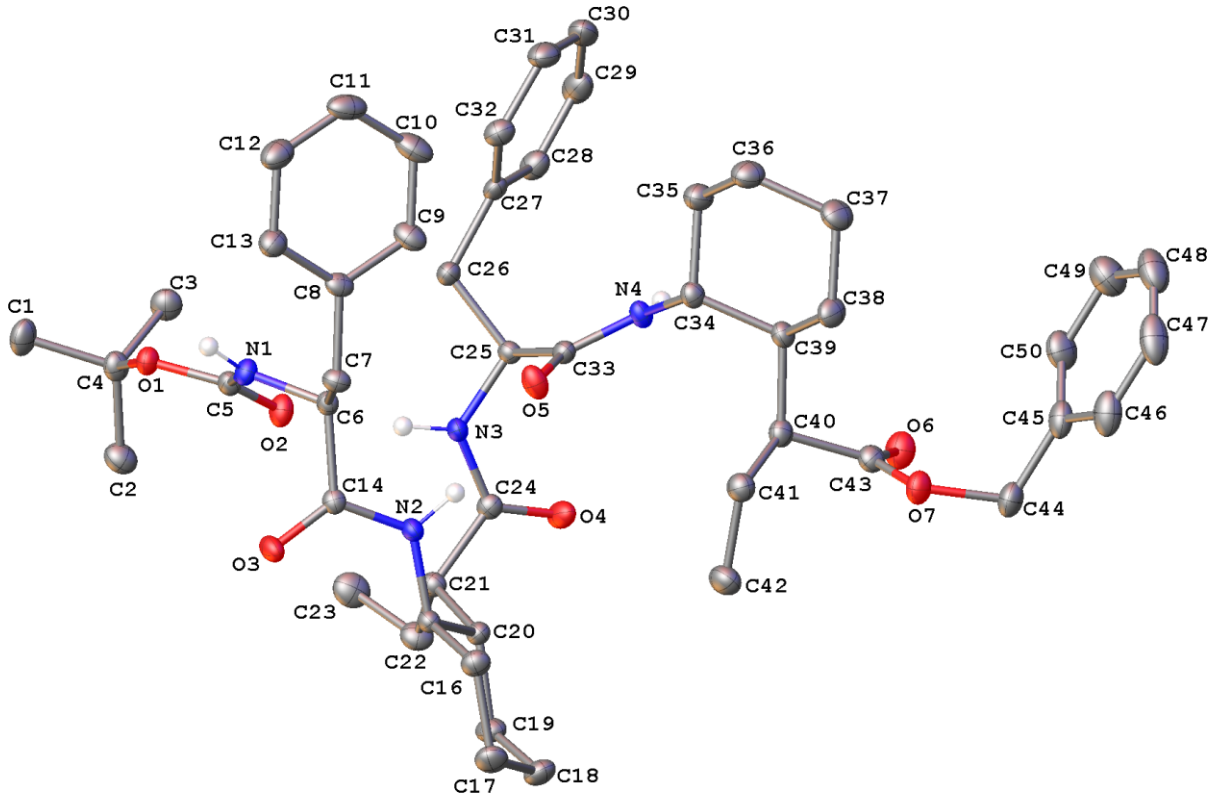


Figure S19. A molecular drawing of the foldamer shown with 50% probability ellipsoids. The disordered atoms, solvent molecules, and non-amido H atoms are omitted.

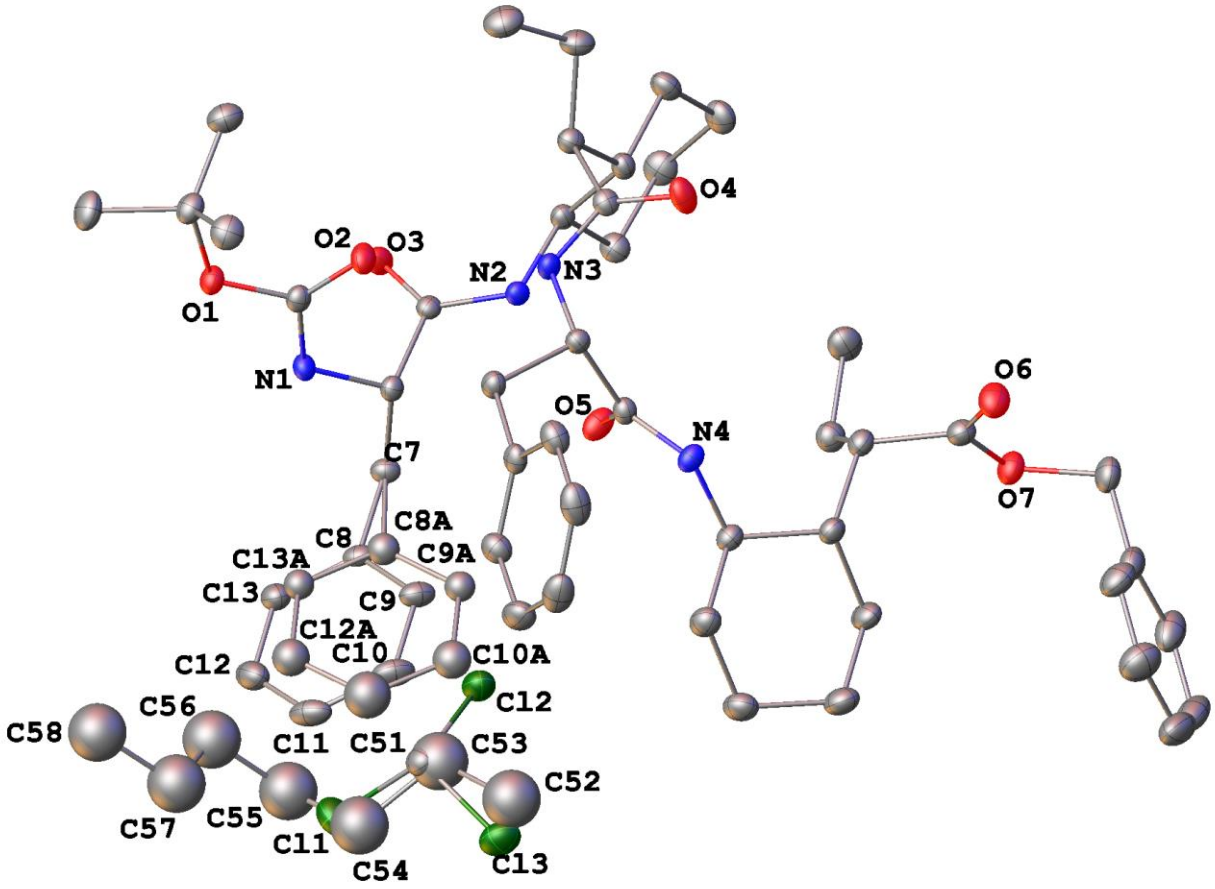


Figure S20. A molecular drawing of Gellman155 shown with 50% probability ellipsoids. The disordered atoms, solvent molecules, and N and O atoms are labeled. All H atoms are omitted. The C51 chloroform and C52-C58 heptane share the same site. Isotropically refined atoms are shown with spheres, anisotropic with ellipsoids.

Table S20. Crystal data and structure refinement for gellman155.	
Identification code	gellman155
Empirical formula	$C_{50}H_{68}N_4O_7$,0.82($CHCl_3$),0.18(C_7H_{16})
Formula weight	952.90
Temperature/K	100.0
Crystal system	triclinic
Space group	$P\bar{1}$
a/Å	11.5447(4)
b/Å	14.2478(5)
c/Å	17.4006(6)
α/°	96.227(2)
β/°	108.4600(18)
γ/°	105.9040(18)
Volume/Å^3	2550.47(16)
Z	2
ρ_{calc}mg/mm^3	1.241
m/mm^{-1}	0.204
F(000)	1020.0
Crystal size/mm^3	0.162 × 0.119 × 0.091
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection	2.526 to 61.23°
Index ranges	$-16 \le h \le 16$, $-20 \le k \le 20$, $-24 \le l \le 24$
Reflections collected	62220
Independent reflections	15641 [R_{int} = 0.0557, R_{sigma} = 0.0599]
Data/restraints/parameters	15641/19/640
Goodness-of-fit on F^2	1.020
Final R indexes [I>=2σ (I)]	R_1 = 0.0546, wR_2 = 0.1260
Final R indexes [all data]	R_1 = 0.0937, wR_2 = 0.1433
Largest diff. peak/hole / e Å^{-3}	0.54/-0.44

Table S21. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for gellman155. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	***x***	***y***	***z***	**U(eq)**
O1	6196.8(11)	3486.4(9)	825.0(7)	17.7(2)
O2	5288.7(12)	3821.7(9)	1763.0(7)	21.0(3)
O3	3369.0(11)	4769.3(9)	384.6(7)	20.3(3)
O4	3356.7(11)	4283.5(10)	3839.7(7)	24.5(3)
O5	5532.1(12)	6488.3(9)	3323.2(7)	22.4(3)
O6	4036.1(13)	7437.3(10)	6276.1(8)	27.6(3)
O7	3573.0(12)	8669.7(9)	5663.8(8)	23.0(3)
N1	6012.3(13)	4985.9(10)	1079.7(8)	16.2(3)
N2	3475.5(12)	5406.4(10)	1665.7(8)	15.0(3)
N3	4779.8(12)	4453.4(10)	3187.1(8)	15.7(3)
N4	6374.2(13)	6735.5(10)	4722.6(8)	16.6(3)
C1	6617.4(19)	2086.5(15)	328.3(12)	28.1(4)
C2	4671.7(18)	1845.9(14)	728.5(12)	26.9(4)

C3	6900.5(18)	2484.6(15)	1829.7(11)	25.4(4)
C4	6077.5(16)	2460.4(13)	946.3(11)	20.0(3)
C5	5790.1(15)	4076.9(12)	1262.9(10)	16.1(3)
C6	5485.4(14)	5671.6(12)	1418.7(10)	14.8(3)
C7	5891.2(15)	6677.9(12)	1174.5(10)	17.8(3)
C8	7333.3(16)	7207.0(14)	1533.0(18)	17.7(4)
C9	7956.6(19)	7575.9(15)	2386.0(13)	23.9(5)
C10	9290(2)	8067.9(16)	2727.9(14)	30.3(5)
C11	10001.9(19)	8196.0(16)	2216.1(15)	30.4(5)
C12	9392.8(19)	7843.0(15)	1372.9(15)	29.1(5)
C13	8062.1(18)	7346.1(14)	1027.0(13)	22.6(5)
C8A	7262(6)	7200(14)	1764(8)	23(9)
C9A	7591(10)	7712(13)	2570(8)	22(5)
C10A	8884(12)	8163(13)	3074(7)	34(6)
C11A	9847(6)	8102(15)	2773(10)	55(11)
C12A	9517(9)	7590(14)	1967(11)	31(6)
C13A	8225(12)	7139(13)	1463(8)	21(5)
C14	4002.5(15)	5231.3(12)	1103(1)	14.6(3)
C15	2079.1(14)	5004.1(12)	1476.3(10)	15.5(3)
C16	1464.6(15)	5819.9(13)	1326.8(11)	20.3(3)
C17	1.5(16)	5405.8(15)	1107.2(12)	25.1(4)
C18	-312.1(16)	4895.6(15)	1771.8(12)	25.3(4)
C19	308.3(15)	4073.8(14)	1904.4(11)	22.0(4)
C20	1778.6(14)	4493.5(12)	2157.8(10)	16.5(3)
C21	2496.9(15)	3713.6(12)	2353.7(10)	16.7(3)
C22	1676.0(17)	2669.0(13)	2360.6(12)	23.9(4)
C23	2385.2(19)	1919.5(14)	2309.1(13)	30.8(4)
C24	3580.7(15)	4161.0(12)	3196.6(10)	16.7(3)
C25	5883.6(14)	5082.9(12)	3899.2(10)	14.8(3)
C26	7125.0(15)	5013.4(13)	3772.6(10)	17.3(3)
C27	8338.9(15)	5566.8(13)	4508.8(10)	17.6(3)
C28	8587.4(17)	5203.9(15)	5233.0(11)	24.0(4)
C29	9693.1(18)	5693.8(17)	5916.0(12)	30.0(4)
C30	10561.7(18)	6568.1(16)	5892.3(12)	30.4(4)
C31	10334.7(17)	6937.9(15)	5179.7(12)	27.8(4)
C32	9227.5(16)	6432.8(14)	4482.5(11)	22.6(4)
C33	5894.4(14)	6166.4(12)	3959.1(10)	16.0(3)
C34	6672.5(15)	7818.7(12)	4868(1)	17.4(3)
C35	8137.1(16)	8336.0(14)	5274.6(12)	24.2(4)
C36	8489.7(17)	9466.1(14)	5395.2(13)	26.9(4)
C37	7788.2(17)	9863.0(14)	5889.9(13)	27.8(4)
C38	6328.9(17)	9333.1(13)	5497.2(12)	23.8(4)
C39	5981.3(15)	8195.7(12)	5392.5(10)	17.8(3)
C40	4497.6(16)	7642.0(13)	5039.2(10)	18.5(3)
C41	3786.7(16)	7837.4(14)	4203.1(11)	22.0(3)
C42	2395.7(17)	7145.1(15)	3804.8(12)	28.4(4)
C43	3998.5(16)	7876.8(13)	5718.6(11)	20.1(3)
C44	3283.6(18)	9047.9(14)	6372.7(12)	26.3(4)
C45	4445.2(19)	9845.9(14)	7002.3(12)	26.0(4)
C46	4545(2)	10846.2(15)	7056.2(13)	33.7(5)
C47	5618(2)	11588.6(16)	7636.8(14)	39.2(5)

C48	6596(2)	11333.5(17)	8163.6(14)	40.5(5)
C49	6503(2)	10342.5(17)	8122.0(14)	40.7(5)
C50	5432(2)	9601.6(15)	7543.0(13)	31.8(4)
Cl1	8802.5(7)	10311.3(6)	883.2(5)	42.4(2)
Cl2	6561.0(5)	9650.4(4)	1341.1(4)	30.44(16)
Cl3	9043.7(6)	10769.6(5)	2600.4(4)	35.00(17)
C51	8242(2)	9888(2)	1647.3(17)	26.0(5)
C52	8620(20)	10399(17)	2625(8)	80
C53	8070(15)	9990(20)	1687(9)	80
C54	9058(11)	10339(10)	1278(8)	80
C55	8516(14)	9988(10)	336(7)	80
C56	8735(11)	9028(10)	36(8)	80
C57	10085(11)	8990(10)	495(10)	80
C58	10289(15)	8028(11)	171(11)	80

Table S22. Anisotropic Displacement Parameters ($Å^2×10^3$) for gellman155. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	**U_{11}**	**U_{22}**	**U_{33}**	**U_{23}**	**U_{13}**	**U_{12}**
O1	21.5(6)	17.1(6)	17.1(6)	3.4(5)	8.7(5)	8.7(5)
O2	28.7(6)	20.9(6)	18.6(6)	6.0(5)	13.6(5)	9.5(5)
O3	18.5(5)	25.1(6)	12.6(6)	0.1(5)	5.2(4)	2.0(5)
O4	19.9(6)	39.6(8)	15.3(6)	6.3(5)	7.8(5)	10.0(5)
O5	31.8(7)	19.4(6)	14.5(6)	3.8(5)	4.6(5)	10.3(5)
O6	37.0(7)	25.1(7)	27.2(7)	9.4(6)	16.9(6)	12.6(6)
O7	29.4(6)	20.7(6)	23.0(6)	3.9(5)	12.0(5)	12.1(5)
N1	19.7(6)	17.2(7)	14.8(7)	4.1(5)	9.5(5)	6.9(5)
N2	14.2(6)	16.6(7)	12.0(6)	1.2(5)	4.1(5)	3.1(5)
N3	15.1(6)	18.1(7)	12.4(6)	0.1(5)	4.6(5)	4.6(5)
N4	20.2(6)	15.5(7)	12.8(6)	1.9(5)	3.2(5)	7.6(5)
C1	36.3(10)	27.7(10)	27.9(10)	4.7(8)	14.4(8)	19.6(8)
C2	26.3(9)	21.3(9)	27.4(10)	2.5(7)	5.7(7)	4.8(7)
C3	27.5(9)	27(1)	23.4(9)	8.5(8)	7.4(7)	12.7(8)
C4	24.8(8)	18.0(8)	19.1(8)	3.9(7)	7.3(7)	10.5(7)
C5	16.0(7)	18.1(8)	13.2(7)	2.2(6)	4.0(6)	6.2(6)
C6	16.0(7)	15.9(8)	12.5(7)	2.4(6)	5.1(6)	5.7(6)
C7	15.5(7)	16.5(8)	18.5(8)	4.1(6)	3.8(6)	3.8(6)
C8	16.4(9)	10.7(9)	21.9(11)	2.4(8)	3.6(8)	2.4(7)
C9	23.2(9)	18.8(10)	22.2(10)	3.5(8)	3.9(8)	0.8(8)
C10	26.5(12)	22.8(11)	26.8(12)	2.6(9)	-1.1(9)	-0.8(8)
C11	17.4(9)	19.4(10)	45.5(14)	3.6(9)	3.4(9)	2.9(8)
C12	23.5(9)	22(1)	42.5(13)	3.0(9)	15.9(9)	5.2(8)
C13	21.2(9)	18.2(9)	26.6(11)	1.5(8)	9.5(8)	3.9(7)
C14	17.0(7)	12.8(7)	14.5(7)	4.3(6)	6.4(6)	4.4(6)
C15	14.3(7)	16.9(8)	13.7(7)	1.3(6)	4.4(6)	4.0(6)
C16	19.1(7)	21.6(9)	21.2(8)	6.7(7)	6.4(6)	8.4(7)
C17	20.3(8)	29.4(10)	25.3(9)	6.8(8)	3.7(7)	12.5(7)
C18	17.3(8)	31.7(10)	28.9(10)	5.9(8)	9.1(7)	10.5(7)
C19	15.6(7)	25.4(9)	23.7(9)	5.5(7)	7.5(7)	4.0(7)

C20	14.3(7)	19.4(8)	15.7(8)	2.3(6)	5.8(6)	5.6(6)
C21	16.2(7)	17.3(8)	15.7(8)	3.8(6)	5.3(6)	4.5(6)
C22	22.2(8)	18.2(9)	28.0(9)	6.0(7)	8.0(7)	2.5(7)
C23	34.2(10)	20.5(9)	35.2(11)	8.2(8)	9.1(8)	8.5(8)
C24	17.4(7)	17.7(8)	16.3(8)	5.7(6)	6.1(6)	6.9(6)
C25	14.2(7)	15.7(8)	13.3(7)	2.6(6)	4.1(6)	4.3(6)
C26	15.6(7)	18.9(8)	17.5(8)	1.8(6)	6.1(6)	6.6(6)
C27	14.9(7)	22.3(8)	18.0(8)	2.9(7)	6.5(6)	9.6(6)
C28	22.8(8)	29.8(10)	24.4(9)	9.1(8)	11.2(7)	12.0(7)
C29	28.4(9)	47.2(13)	20.0(9)	9.6(9)	8.8(7)	19.7(9)
C30	20.4(8)	40.9(12)	23.7(9)	-3.9(8)	1.3(7)	12.4(8)
C31	17.0(8)	26.6(10)	35.3(11)	0.4(8)	8.5(7)	3.9(7)
C32	19.2(8)	25.0(9)	25.4(9)	7.3(7)	9.0(7)	8.4(7)
C33	14.6(7)	19.1(8)	14.5(7)	2.7(6)	5.3(6)	6.3(6)
C34	19.2(7)	15.2(8)	17.0(8)	2.3(6)	5.6(6)	6.2(6)
C35	19.3(8)	23.1(9)	26.7(9)	0.9(7)	6.1(7)	6.1(7)
C36	19.6(8)	23.3(9)	33.1(10)	2.9(8)	7.8(7)	3.1(7)
C37	26.0(9)	18.2(9)	32.4(10)	-0.9(8)	7.7(8)	2.7(7)
C38	23.9(8)	16.0(8)	30.7(10)	0.1(7)	10.7(7)	6.0(7)
C39	18.6(7)	16.2(8)	16.5(8)	0.5(6)	5.3(6)	4.8(6)
C40	21.0(8)	15.7(8)	19.3(8)	2.1(6)	8.2(6)	6.3(6)
C41	22.2(8)	23.5(9)	20.4(9)	4.7(7)	7.9(7)	7.4(7)
C42	24.2(9)	34.1(11)	21.4(9)	2.7(8)	4.4(7)	6.7(8)
C43	19.8(7)	17.0(8)	21.5(9)	1.9(7)	6.9(6)	4.4(6)
C44	32.0(9)	26.4(10)	26.7(10)	3.5(8)	15.5(8)	14.2(8)
C45	37.6(10)	22.3(9)	24.3(9)	4.1(7)	19.0(8)	10.4(8)
C46	55.1(13)	28.6(11)	29.0(11)	10.3(9)	24.3(10)	19.2(10)
C47	68.5(15)	18.5(10)	35.9(12)	4.2(9)	31.1(11)	8.9(10)
C48	51.8(13)	27.7(11)	33.7(12)	-4.6(9)	20.2(10)	-1.1(10)
C49	43.6(12)	35.8(12)	33.8(12)	0.9(10)	8.3(10)	8.6(10)
C50	41.2(11)	20.7(9)	31.7(11)	1.1(8)	12.6(9)	9.7(8)
Cl1	42.8(4)	45.0(4)	44.2(4)	13.1(3)	26.0(3)	8.4(3)
Cl2	24.4(3)	28.3(3)	32.5(3)	-2.6(2)	10.4(2)	2.7(2)
Cl3	28.8(3)	31.6(4)	33.5(3)	-0.2(3)	3.3(2)	5.0(2)
C51	25.2(11)	21.6(12)	30.5(12)	2.4(9)	10.8(9)	7.1(9)

Table S23. Bond Lengths for gellman155.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
O1	C4	1.475(2)	C19	C20	1.532(2)
O1	C5	1.3464(19)	C20	C21	1.564(2)
O2	C5	1.2235(19)	C21	C22	1.533(2)
O3	C14	1.2287(19)	C21	C24	1.523(2)
O4	C24	1.231(2)	C22	C23	1.524(3)
O5	C33	1.2377(19)	C25	C26	1.544(2)
O6	C43	1.206(2)	C25	C33	1.532(2)
O7	C43	1.351(2)	C26	C27	1.505(2)
O7	C44	1.463(2)	C27	C28	1.390(2)
N1	C5	1.343(2)	C27	C32	1.387(2)

N1	C6	1.449(2)	C28	C29	1.378(3)
N2	C14	1.341(2)	C29	C30	1.383(3)
N2	C15	1.4670(19)	C30	C31	1.377(3)
N3	C24	1.338(2)	C31	C32	1.396(3)
N3	C25	1.448(2)	C34	C35	1.533(2)
N4	C33	1.335(2)	C34	C39	1.532(2)
N4	C34	1.461(2)	C35	C36	1.522(3)
C1	C4	1.520(2)	C36	C37	1.518(3)
C2	C4	1.518(2)	C37	C38	1.529(2)
C3	C4	1.521(2)	C38	C39	1.534(2)
C6	C7	1.532(2)	C39	C40	1.556(2)
C6	C14	1.539(2)	C40	C41	1.525(2)
C7	C8	1.510(2)	C40	C43	1.514(2)
C7	C8A	1.507(4)	C41	C42	1.520(2)
C8	C9	1.394(3)	C44	C45	1.502(3)
C8	C13	1.392(3)	C45	C46	1.388(3)
C9	C10	1.398(3)	C45	C50	1.383(3)
C10	C11	1.385(3)	C46	C47	1.390(3)
C11	C12	1.376(3)	C47	C48	1.374(3)
C12	C13	1.396(3)	C48	C49	1.379(3)
C8A	C9A	1.3900	C49	C50	1.387(3)
C8A	C13A	1.3900	Cl1	C51	1.747(3)
C9A	C10A	1.3900	Cl2	C51	1.764(2)
C10A	C11A	1.3900	Cl3	C51	1.767(3)
C11A	C12A	1.3900	C52	C53	1.532(3)
C12A	C13A	1.3900	C53	C54	1.528(3)
C15	C16	1.524(2)	C54	C55	1.526(3)
C15	C20	1.538(2)	C55	C56	1.526(3)
C16	C17	1.531(2)	C56	C57	1.528(3)
C17	C18	1.517(3)	C57	C58	1.532(3)
C18	C19	1.533(2)			

Table S24. Bond Angles for gellman155.

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C5	O1	C4	120.05(13)	O4	C24	N3	122.40(15)
C43	O7	C44	116.04(14)	O4	C24	C21	121.73(14)
C5	N1	C6	117.88(13)	N3	C24	C21	115.80(14)
C14	N2	C15	121.77(13)	N3	C25	C26	108.46(13)
C24	N3	C25	122.49(13)	N3	C25	C33	109.02(12)
C33	N4	C34	122.00(14)	C33	C25	C26	109.05(13)
O1	C4	C1	101.54(14)	C27	C26	C25	113.46(13)
O1	C4	C2	110.72(13)	C28	C27	C26	119.62(16)
O1	C4	C3	110.12(14)	C32	C27	C26	121.56(15)
C1	C4	C3	111.13(14)	C32	C27	C28	118.82(16)
C2	C4	C1	111.00(15)	C29	C28	C27	120.87(18)
C2	C4	C3	111.88(15)	C28	C29	C30	120.07(18)
O2	C5	O1	124.59(15)	C31	C30	C29	119.92(18)
O2	C5	N1	124.45(15)	C30	C31	C32	120.07(18)

N1	C5	O1	110.96(14)	C27	C32	C31	120.23(17)
N1	C6	C7	110.09(13)	O5	C33	N4	123.20(15)
N1	C6	C14	110.22(13)	O5	C33	C25	120.45(14)
C7	C6	C14	110.04(12)	N4	C33	C25	116.25(14)
C8	C7	C6	113.27(14)	N4	C34	C35	109.93(13)
C8A	C7	C6	104.8(8)	N4	C34	C39	112.84(14)
C9	C8	C7	119.9(2)	C39	C34	C35	110.41(14)
C13	C8	C7	121.3(2)	C36	C35	C34	111.41(14)
C13	C8	C9	118.76(17)	C37	C36	C35	111.20(16)
C8	C9	C10	120.6(2)	C36	C37	C38	111.20(15)
C11	C10	C9	119.9(2)	C37	C38	C39	111.87(14)
C12	C11	C10	119.88(19)	C34	C39	C38	109.15(14)
C11	C12	C13	120.6(2)	C34	C39	C40	112.57(13)
C8	C13	C12	120.24(19)	C38	C39	C40	112.92(13)
C9A	C8A	C7	123.9(10)	C41	C40	C39	114.37(14)
C9A	C8A	C13A	120.0	C43	C40	C39	106.80(13)
C13A	C8A	C7	116.1(10)	C43	C40	C41	115.00(14)
C10A	C9A	C8A	120.0	C42	C41	C40	112.79(15)
C9A	C10A	C11A	120.0	O6	C43	O7	123.20(16)
C12A	C11A	C10A	120.0	O6	C43	C40	123.77(15)
C11A	C12A	C13A	120.0	O7	C43	C40	112.92(14)
C12A	C13A	C8A	120.0	O7	C44	C45	111.31(15)
O3	C14	N2	123.66(14)	C46	C45	C44	120.47(18)
O3	C14	C6	121.51(14)	C50	C45	C44	120.88(17)
N2	C14	C6	114.82(13)	C50	C45	C46	118.65(19)
N2	C15	C16	110.52(13)	C45	C46	C47	120.8(2)
N2	C15	C20	111.49(12)	C48	C47	C46	119.8(2)
C16	C15	C20	111.81(13)	C47	C48	C49	120.0(2)
C15	C16	C17	111.15(14)	C48	C49	C50	120.2(2)
C18	C17	C16	110.74(14)	C45	C50	C49	120.6(2)
C17	C18	C19	110.50(14)	Cl1	C51	Cl2	110.76(16)
C20	C19	C18	111.53(14)	Cl1	C51	Cl3	110.93(15)
C15	C20	C21	111.46(12)	Cl2	C51	Cl3	109.53(13)
C19	C20	C15	108.23(13)	C54	C53	C52	112.8(4)
C19	C20	C21	114.84(14)	C55	C54	C53	113.9(4)
C22	C21	C20	116.68(13)	C54	C55	C56	113.7(4)
C24	C21	C20	107.03(13)	C55	C56	C57	114.0(4)
C24	C21	C22	109.24(14)	C56	C57	C58	113.0(4)
C23	C22	C21	110.69(14)				

Table S25. Hydrogen Bonds for gellman155.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
N1	H1	O3[1]	0.871(9)	2.018(10)	2.8868(18)	175.9(18)
N2	H2	O5	0.870(9)	2.141(10)	3.0042(18)	171.3(18)
N3	H3	O2	0.875(9)	1.966(10)	2.8326(18)	170.5(18)
N4	H4	O4[2]	0.867(9)	2.144(10)	3.0010(19)	169.4(18)

[1]1-X,1-Y,-Z; [2]1-X,1-Y,1-Z

Table S26. Torsion Angles for gellman155.

A	B	C	D	Angle/°	A	B	C	D	Angle/°
O7	C44	C45	C46	102.51(19)	C18	C19	C20	C21	176.77(14)
O7	C44	C45	C50	-77.9(2)	C19	C20	C21	C22	-6.4(2)
N1	C6	C7	C8	-63.9(2)	C19	C20	C21	C24	-129.01(15)
N1	C6	C7	C8A	-79.1(6)	C20	C15	C16	C17	-56.57(18)
N1	C6	C14	O3	-39.4(2)	C20	C21	C22	C23	163.66(15)
N1	C6	C14	N2	141.55(14)	C20	C21	C24	O4	68.28(19)
N2	C15	C16	C17	178.60(14)	C20	C21	C24	N3	-108.69(15)
N2	C15	C20	C19	-178.66(13)	C22	C21	C24	O4	-58.9(2)
N2	C15	C20	C21	-51.45(17)	C22	C21	C24	N3	124.13(15)
N3	C25	C26	C27	-174.90(13)	C24	N3	C25	C26	163.20(15)
N3	C25	C33	O5	-35.47(19)	C24	N3	C25	C33	-78.17(18)
N3	C25	C33	N4	147.96(14)	C24	C21	C22	C23	-74.83(18)
N4	C34	C35	C36	177.07(14)	C25	N3	C24	O4	-10.7(2)
N4	C34	C39	C38	-178.84(13)	C25	N3	C24	C21	166.20(14)
N4	C34	C39	C40	-52.63(18)	C25	C26	C27	C28	70.3(2)
C4	O1	C5	O2	-1.4(2)	C25	C26	C27	C32	-110.12(17)
C4	O1	C5	N1	178.39(13)	C26	C25	C33	O5	82.79(18)
C5	O1	C4	C1	178.91(13)	C26	C25	C33	N4	-93.79(16)
C5	O1	C4	C2	60.99(19)	C26	C27	C28	C29	179.82(16)
C5	O1	C4	C3	-63.26(18)	C26	C27	C32	C31	179.01(15)
C5	N1	C6	C7	177.45(13)	C27	C28	C29	C30	1.2(3)
C5	N1	C6	C14	-60.98(18)	C28	C27	C32	C31	-1.4(2)
C6	N1	C5	O1	172.37(12)	C28	C29	C30	C31	-1.4(3)
C6	N1	C5	O2	-7.8(2)	C29	C30	C31	C32	0.2(3)
C6	C7	C8	C9	-67.9(2)	C30	C31	C32	C27	1.2(3)
C6	C7	C8	C13	112.8(2)	C32	C27	C28	C29	0.2(2)
C6	C7	C8A	C9A	-81.3(12)	C33	N4	C34	C35	-112.07(17)
C6	C7	C8A	C13A	98.1(10)	C33	N4	C34	C39	124.20(16)
C7	C6	C14	O3	82.19(18)	C33	C25	C26	C27	66.49(17)
C7	C6	C14	N2	-96.84(16)	C34	N4	C33	O5	-8.4(2)
C7	C8	C9	C10	-179.95(18)	C34	N4	C33	C25	168.07(13)
C7	C8	C13	C12	179.61(17)	C34	C35	C36	C37	55.6(2)
C7	C8A	C9A	C10A	179.3(18)	C34	C39	C40	C41	-66.47(18)
C7	C8A	C13A	C12A	-179.4(17)	C34	C39	C40	C43	165.11(14)
C8	C7	C8A	C9A	156(4)	C35	C34	C39	C38	57.69(18)
C8	C7	C8A	C13A	-25(2)	C35	C34	C39	C40	-176.09(14)
C8	C9	C10	C11	0.3(3)	C35	C36	C37	C38	-54.1(2)
C9	C8	C13	C12	0.3(3)	C36	C37	C38	C39	55.7(2)
C9	C10	C11	C12	0.3(3)	C37	C38	C39	C34	-57.18(19)
C10	C11	C12	C13	-0.7(3)	C37	C38	C39	C40	176.81(15)
C11	C12	C13	C8	0.4(3)	C38	C39	C40	C41	57.69(19)
C13	C8	C9	C10	-0.6(3)	C38	C39	C40	C43	-70.73(18)
C8A	C7	C8	C9	-6(3)	C39	C34	C35	C36	-57.79(19)
C8A	C7	C8	C13	175(3)	C39	C40	C41	C42	169.46(14)
C8A	C9A	C10A	C11A	0.0	C39	C40	C43	O6	-82.8(2)
C9A	C8A	C13A	C12A	0.0	C39	C40	C43	O7	93.59(16)
C9A	C10A	C11A	C12A	0.0	C41	C40	C43	O6	149.14(17)

C10A	C11A	C12A	C13A	0.0	C41	C40	C43	O7	-34.5(2)
C11A	C12A	C13A	C8A	0.0	C43	O7	C44	C45	90.81(18)
C13A	C8A	C9A	C10A	0.0	C43	C40	C41	C42	-66.4(2)
C14	N2	C15	C16	-103.71(17)	C44	O7	C43	O6	6.5(2)
C14	N2	C15	C20	131.28(15)	C44	O7	C43	C40	-169.93(14)
C14	C6	C7	C8	174.41(17)	C44	C45	C46	C47	-179.93(18)
C14	C6	C7	C8A	159.2(6)	C44	C45	C50	C49	179.86(19)
C15	N2	C14	O3	3.5(2)	C45	C46	C47	C48	0.2(3)
C15	N2	C14	C6	-177.45(13)	C46	C45	C50	C49	-0.6(3)
C15	C16	C17	C18	55.3(2)	C46	C47	C48	C49	-0.8(3)
C15	C20	C21	C22	-129.89(15)	C47	C48	C49	C50	0.7(3)
C15	C20	C21	C24	107.46(15)	C48	C49	C50	C45	0.0(3)
C16	C15	C20	C19	57.05(17)	C50	C45	C46	C47	0.5(3)
C16	C15	C20	C21	-175.74(13)	C52	C53	C54	C55	-176.9(17)
C16	C17	C18	C19	-56.1(2)	C53	C54	C55	C56	-94.7(19)
C17	C18	C19	C20	58.7(2)	C54	C55	C56	C57	-42.6(17)
C18	C19	C20	C15	-58.00(18)	C55	C56	C57	C58	-178.9(14)

Table S27. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for gellman155.

Atom	***x***	***y***	***z***	**U(eq)**
H1	6239(18)	5075(14)	654(9)	19
H2	4007(15)	5701(13)	2169(7)	18
H3	4906(18)	4313(14)	2724(8)	19
H4	6559(18)	6463(14)	5148(9)	20
H1A	6105	2121	-231	42
H1B	6577	1393	343	42
H1C	7518	2504	474	42
H2A	4356	2083	1147	40
H2B	4596	1142	714	40
H2C	4155	1916	183	40
H3A	7789	2916	1957	38
H3B	6895	1807	1886	38
H3C	6548	2747	2216	38
H6	5832	5779	2037	18
H7AA	5594	6568	562	21
H7AB	5457	7114	1366	21
H7BC	5849	6580	593	21
H7BD	5325	7069	1236	21
H9	7470	7492	2738	29
H10	9707	8314	3310	36
H11	10909	8527	2447	36
H12	9882	7938	1023	35
H13	7652	7102	444	27
H9A	6933	7754	2777	27
H10A	9109	8514	3625	41
H11A	10730	8411	3117	66
H12A	10175	7548	1760	38

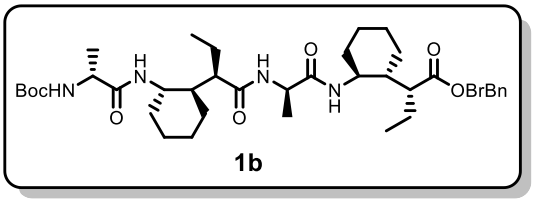
H13A	7999	6788	912	25
H15	1697	4484	950	19
H16A	1646	6109	868	24
H16B	1851	6360	1832	24
H17A	-365	5958	1051	30
H17B	-399	4921	568	30
H18A	-1261	4600	1604	30
H18B	13	5395	2298	30
H19A	97	3752	2342	26
H19B	-55	3557	1385	26
H20	2112	5024	2674	20
H21	2901	3638	1930	20
H22A	1483	2691	2876	29
H22B	846	2453	1884	29
H23A	2480	1835	1768	46
H23B	1890	1276	2376	46
H23C	3242	2166	2750	46
H25	5836	4858	4416	18
H26A	7060	4301	3655	21
H26B	7188	5286	3282	21
H28	7987	4610	5258	29
H29	9858	5431	6404	36
H30	11314	6913	6367	36
H31	10932	7537	5162	33
H32	9082	6683	3988	27
H34	6377	8002	4317	21
H35A	8455	8158	5819	29
H35B	8566	8095	4922	29
H36A	9434	9779	5689	32
H36B	8260	9649	4847	32
H37A	7975	10589	5916	33
H37B	8108	9763	6464	33
H38A	5996	9496	4948	29
H38B	5905	9580	5851	29
H39	6331	8056	5956	21
H40	4374	6910	4951	22
H41A	3794	8538	4280	26
H41B	4252	7752	3824	26
H42A	1989	7294	3267	43
H42B	1920	7245	4167	43
H42C	2381	6450	3724	43
H44A	2996	8490	6635	32
H44B	2569	9324	6178	32
H46	3872	11025	6692	40
H47	5675	12271	7669	47
H48	7335	11840	8556	49
H49	7175	10167	8491	49
H50	5375	8920	7518	38
H51	8423	9251	1728	31
H52A	8521	11055	2742	120
H52B	9538	10464	2845	120

H52C	8152	9940	2890	120
H53A	7764	9250	1576	96
H53B	7316	10206	1433	96
H54A	9401	11078	1418	96
H54B	9790	10092	1512	96
H55A	8922	10523	92	96
H55B	7576	9880	131	96
H56A	8589	8953	-563	96
H56B	8088	8453	102	96
H57A	10227	9048	1092	96
H57B	10735	9570	438	96
H58A	10807	8166	-179	120
H58B	9449	7523	-156	120
H58C	10741	7778	640	120

Table S28 Atomic Occupancy for gellman155.

Atom	***Occupancy***	**Atom**	***Occupancy***	**Atom**	***Occupancy***
H7AA	0.919(4)	H7AB	0.919(4)	H7BC	0.081(4)
H7BD	0.081(4)	C8	0.919(4)	C9	0.919(4)
H9	0.919(4)	C10	0.919(4)	H10	0.919(4)
C11	0.919(4)	H11	0.919(4)	C12	0.919(4)
H12	0.919(4)	C13	0.919(4)	H13	0.919(4)
C8A	0.081(4)	C9A	0.081(4)	H9A	0.081(4)
C10A	0.081(4)	H10A	0.081(4)	C11A	0.081(4)
H11A	0.081(4)	C12A	0.081(4)	H12A	0.081(4)
C13A	0.081(4)	H13A	0.081(4)	Cl1	0.8161(15)
Cl2	0.8161(15)	Cl3	0.8161(15)	C51	0.8161(15)
H51	0.8161(15)	C52	0.1839(15)	H52A	0.1839(15)
H52B	0.1839(15)	H52C	0.1839(15)	C53	0.1839(15)
H53A	0.1839(15)	H53B	0.1839(15)	C54	0.1839(15)
H54A	0.1839(15)	H54B	0.1839(15)	C55	0.1839(15)
H55A	0.1839(15)	H55B	0.1839(15)	C56	0.1839(15)
H56A	0.1839(15)	H56B	0.1839(15)	C57	0.1839(15)
H57A	0.1839(15)	H57B	0.1839(15)	C58	0.1839(15)
H58A	0.1839(15)	H58B	0.1839(15)	H58C	0.1839(15)

Crystallographic Experimental Section for Compound 1b



1b

Data Collection

A colorless crystal with approximate dimensions 0.50 x 0.12 x 0.11 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker SMART APEXII diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 41 frames collected at intervals of 0.6º in a 25º range about ω with the exposure time of 5 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9956 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.82 Å. A total of 98960 data were harvested by collecting 19 sets of frames with 0.7º scans in ω with an exposure time 20-60 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were consistent for the space group $P6_1$ that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms were refined with anisotropic displacement coefficients. All hydrogen atoms (except amido H atoms) were included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

There was one partially occupied diatomic solvate molecule of methanol (or two water molecules) present in the asymmetric unit. A significant amount of time was invested in identifying and refining the disordered molecules. Bond length restraints were applied to model the molecules but the resulting isotropic displacement coefficients suggested the molecules were mobile. In addition, the refinement was computationally unstable. Option SQUEEZE of program PLATON [5] was used to correct the diffraction data for diffuse scattering effects and to identify

the solvate molecule. PLATON calculated the upper limit of volume that can be occupied by the solvent to be 202 $Å^3$, or 3% of the unit cell volume. The program calculated 54 electrons in the unit cell for the diffuse species. This approximately corresponds to one half of methanol molecule per foldamer in the asymmetric unit (54 electrons). Please note that all derived results in the following tables are based on the known contents. No data are given for the diffusely scattering species.

The final least-squares refinement of 459 parameters against 7816 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0410 and 0.1083, respectively. The final difference Fourier map was featureless.

The molecular diagram is drawn with 40% probability ellipsoids.

References

[1] Bruker-AXS. (2007) APEX2, SADABS, and SAINT Software Reference Manuals. Bruker-AXS, Madison, Wisconsin, USA.

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[4] Guzei, I.A. (2006-2008). Internal laboratory computer programs "Inserter", "FCF_filter", "Modicifer".

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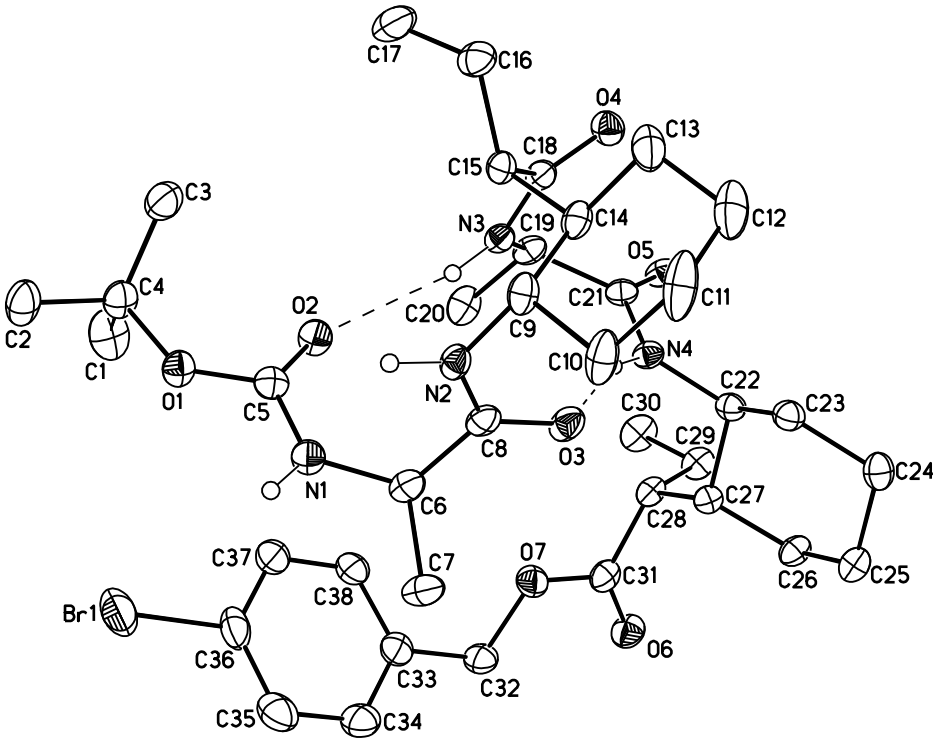


Figure S21. A molecular drawing of Gellman126. All non-amido H atoms are omitted. Two hydrogen bonds are shown with dashed lines.

Table S29. Crystal data and structure refinement for gellman126.

Identification code	**gellman126**	
Empirical formula	C_{38} H_{59} Br N_4 O_7 .1/2 MeOH	
Formula weight	763.80	
Temperature	100(2) K	
Wavelength	1.54178 Å	
Crystal system	Hexagonal	
Space group	$P6_1$	
Unit cell dimensions	a = 25.5011(4) Å	α= 90°.
	b = 25.5011(4) Å	β= 90°.
	c = 11.0911(2) Å	γ = 120°.
Volume	6246.30(18) $Å^3$	
Z	6	
Density (calculated)	1.218 Mg/m^3	
Absorption coefficient	1.736 mm^{-1}	
F(000)	2436	
Crystal size	0.50 x 0.12 x 0.11 mm^3	
Theta range for data collection	2.00 to 71.66°.	
Index ranges	-30<=h<=30, -31<=k<=30, -12<=l<=13	
Reflections collected	98960	
Independent reflections	7816 [R(int) = 0.0658]	
Completeness to theta = 67.00°	99.8 %	
Absorption correction	Empirical with SADABS	
Max. and min. transmission	0.8374 and 0.4797	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	7816 / 1 / 459	
Goodness-of-fit on F^2	1.046	
Final R indices [I>2sigma(I)]	R1 = 0.0410, wR2 = 0.1074	
R indices (all data)	R1 = 0.0425, wR2 = 0.1083	
Absolute structure parameter (Flack x)	-0.008(12)	
Absolute structure parameter (Hooft y)	-0.012(3)	
Largest diff. peak and hole	0.678 and -0.379 $e.Å^{-3}$	

Table S30. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for gellman126. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Br(1)	5112(1)	5276(1)	7996(1)	65(1)
O(1)	6534(1)	6760(1)	8051(2)	28(1)
O(2)	6280(1)	7410(1)	7146(2)	31(1)
O(3)	6362(1)	7484(1)	3522(2)	32(1)
O(4)	7314(1)	9411(1)	4738(2)	30(1)
O(5)	5732(1)	9098(1)	4231(2)	30(1)
O(6)	3548(1)	6853(1)	3166(2)	37(1)
O(7)	4249(1)	6788(1)	4274(2)	33(1)
N(1)	6226(1)	6600(1)	6166(2)	29(1)
N(2)	7081(1)	7395(1)	4574(2)	26(1)
N(3)	6625(1)	8543(1)	5636(2)	24(1)
N(4)	5894(1)	8352(1)	3558(2)	24(1)
C(1)	6046(2)	6908(2)	9793(3)	50(1)
C(2)	6902(2)	6688(1)	9918(3)	43(1)

C(3)	7111(1)	7706(1)	9184(3)	45(1)
C(4)	6648(1)	7035(1)	9243(2)	32(1)
C(5)	6341(1)	6962(1)	7128(2)	27(1)
C(6)	6013(1)	6725(1)	5050(2)	30(1)
C(7)	5731(1)	6158(1)	4263(3)	43(1)
C(8)	6505(1)	7243(1)	4327(2)	27(1)
C(9)	7611(1)	7853(1)	3928(2)	29(1)
C(10)	7612(1)	7635(1)	2638(2)	40(1)
C(11)	8168(2)	8092(2)	1929(3)	52(1)
C(12)	8215(1)	8708(2)	1939(3)	48(1)
C(13)	8239(1)	8925(1)	3230(2)	39(1)
C(14)	7663(1)	8479(1)	3929(2)	28(1)
C(15)	7618(1)	8678(1)	5231(2)	25(1)
C(16)	8214(1)	9148(1)	5819(2)	34(1)
C(17)	8135(1)	9236(1)	7149(3)	45(1)
C(18)	7178(1)	8920(1)	5193(2)	25(1)
C(19)	6157(1)	8717(1)	5646(2)	27(1)
C(20)	5631(1)	8274(1)	6416(2)	37(1)
C(21)	5922(1)	8747(1)	4388(2)	24(1)
C(22)	5610(1)	8294(1)	2381(2)	24(1)
C(23)	6008(1)	8264(1)	1392(2)	28(1)
C(24)	5727(1)	8204(1)	139(2)	31(1)
C(25)	5088(1)	7654(1)	91(2)	30(1)
C(26)	4693(1)	7687(1)	1086(2)	28(1)
C(27)	4970(1)	7734(1)	2339(2)	24(1)
C(28)	4558(1)	7733(1)	3371(2)	26(1)
C(29)	4308(1)	8164(1)	3199(2)	32(1)
C(30)	4082(1)	8285(1)	4381(3)	41(1)
C(31)	4053(1)	7086(1)	3575(2)	30(1)
C(32)	3811(1)	6168(1)	4558(2)	34(1)
C(33)	4123(1)	5941(1)	5405(2)	34(1)
C(34)	4063(1)	5376(1)	5261(3)	41(1)
C(35)	4344(1)	5170(1)	6049(3)	46(1)
C(36)	4697(1)	5543(1)	6956(3)	44(1)
C(37)	4764(1)	6105(1)	7124(3)	40(1)
C(38)	4470(1)	6304(1)	6353(3)	36(1)

Table S31. Bond lengths [Å] and angles [°] for gellman126.

Br(1)-C(36)	1.904(3)	C(10)-H(10A)	0.9900
O(1)-C(5)	1.345(3)	C(11)-C(12)	1.514(5)
O(1)-C(4)	1.458(3)	C(11)-H(11B)	0.9900
O(2)-C(5)	1.228(3)	C(11)-H(11A)	0.9900
O(3)-C(8)	1.239(3)	C(12)-C(13)	1.526(4)
O(4)-C(18)	1.229(3)	C(12)-H(12A)	0.9900
O(5)-C(21)	1.225(3)	C(12)-H(12B)	0.9900
O(6)-C(31)	1.205(3)	C(13)-C(14)	1.543(3)
O(7)-C(31)	1.345(3)	C(13)-H(13B)	0.9900
O(7)-C(32)	1.444(3)	C(13)-H(13A)	0.9900
N(1)-C(5)	1.343(3)	C(14)-C(15)	1.554(3)
N(1)-C(6)	1.449(3)	C(14)-H(14)	1.0000
N(1)-H(1)	0.8800	C(15)-C(18)	1.526(3)
N(2)-C(8)	1.344(3)	C(15)-C(16)	1.533(3)
N(2)-C(9)	1.458(3)	C(15)-H(15)	1.0000
N(2)-H(2)	0.8800	C(16)-C(17)	1.521(4)
N(3)-C(18)	1.342(3)	C(16)-H(16A)	0.9900
N(3)-C(19)	1.466(3)	C(16)-H(16B)	0.9900
N(3)-H(3)	0.8800	C(17)-H(17B)	0.9800
N(4)-C(21)	1.338(3)	C(17)-H(17C)	0.9800
N(4)-C(22)	1.464(3)	C(17)-H(17A)	0.9800
N(4)-H(4)	0.8800	C(19)-C(20)	1.514(3)
C(1)-C(4)	1.527(4)	C(19)-C(21)	1.535(3)
C(1)-H(1C)	0.9800	C(19)-H(19)	1.0000
C(1)-H(1A)	0.9800	C(20)-H(20B)	0.9800
C(1)-H(1B)	0.9800	C(20)-H(20C)	0.9800
C(2)-C(4)	1.528(4)	C(20)-H(20A)	0.9800
C(2)-H(2C)	0.9800	C(22)-C(23)	1.522(3)
C(2)-H(2B)	0.9800	C(22)-C(27)	1.541(3)
C(2)-H(2A)	0.9800	C(22)-H(22)	1.0000
C(3)-C(4)	1.518(4)	C(23)-C(24)	1.535(3)
C(3)-H(3A)	0.9800	C(23)-H(23A)	0.9900
C(3)-H(3B)	0.9800	C(23)-H(23B)	0.9900
C(3)-H(3C)	0.9800	C(24)-C(25)	1.529(3)
C(6)-C(8)	1.518(3)	C(24)-H(24B)	0.9900
C(6)-C(7)	1.525(3)	C(24)-H(24A)	0.9900
C(6)-H(6)	1.0000	C(25)-C(26)	1.525(3)
C(7)-H(7B)	0.9800	C(25)-H(25B)	0.9900
C(7)-H(7A)	0.9800	C(25)-H(25A)	0.9900
C(7)-H(7C)	0.9800	C(26)-C(27)	1.535(3)
C(9)-C(14)	1.535(3)	C(26)-H(26A)	0.9900
C(9)-C(10)	1.536(3)	C(26)-H(26B)	0.9900
C(9)-H(9)	1.0000	C(27)-C(28)	1.552(3)
C(10)-C(11)	1.527(4)	C(27)-H(27)	1.0000
C(10)-H(10B)	0.9900	C(28)-C(31)	1.518(3)

C(28)-C(29)	1.531(3)	C(33)-C(34)	1.379(4)
C(28)-H(28)	1.0000	C(33)-C(38)	1.389(4)
C(29)-C(30)	1.526(4)	C(34)-C(35)	1.389(4)
C(29)-H(29B)	0.9900	C(34)-H(34)	0.9500
C(29)-H(29A)	0.9900	C(35)-C(36)	1.368(5)
C(30)-H(30B)	0.9800	C(35)-H(35)	0.9500
C(30)-H(30C)	0.9800	C(36)-C(37)	1.370(4)
C(30)-H(30A)	0.9800	C(37)-C(38)	1.390(4)
C(32)-C(33)	1.519(4)	C(37)-H(37)	0.9500
C(32)-H(32A)	0.9900	C(38)-H(38)	0.9500
C(32)-H(32B)	0.9900		

C(5)-O(1)-C(4)	*121.91(17)*	C(3)-C(4)-C(1)	113.0(2)
C(31)-O(7)-C(32)	*116.27(18)*	O(1)-C(4)-C(2)	101.81(18)
C(5)-N(1)-C(6)	*120.37(19)*	C(3)-C(4)-C(2)	110.9(2)
C(5)-N(1)-H(1)	*119.8*	C(1)-C(4)-C(2)	110.7(2)
C(6)-N(1)-H(1)	*119.8*	O(2)-C(5)-N(1)	124.5(2)
C(8)-N(2)-C(9)	*124.96(19)*	O(2)-C(5)-O(1)	125.8(2)
C(8)-N(2)-H(2)	*117.5*	N(1)-C(5)-O(1)	109.72(19)
C(9)-N(2)-H(2)	*117.5*	N(1)-C(6)-C(8)	113.86(19)
C(18)-N(3)-C(19)	*120.65(18)*	N(1)-C(6)-C(7)	109.4(2)
C(18)-N(3)-H(3)	*119.7*	C(8)-C(6)-C(7)	108.4(2)
C(19)-N(3)-H(3)	*119.7*	N(1)-C(6)-H(6)	108.3
C(21)-N(4)-C(22)	*121.82(18)*	C(8)-C(6)-H(6)	108.3
C(21)-N(4)-H(4)	*119.1*	C(7)-C(6)-H(6)	108.3
C(22)-N(4)-H(4)	*119.1*	C(6)-C(7)-H(7B)	109.5
C(4)-C(1)-H(1C)	*109.5*	C(6)-C(7)-H(7A)	109.5
C(4)-C(1)-H(1A)	*109.5*	H(7B)-C(7)-H(7A)	109.5
H(1C)-C(1)-H(1A)	*109.5*	C(6)-C(7)-H(7C)	109.5
C(4)-C(1)-H(1B)	*109.5*	H(7B)-C(7)-H(7C)	109.5
H(1C)-C(1)-H(1B)	*109.5*	H(7A)-C(7)-H(7C)	109.5
H(1A)-C(1)-H(1B)	*109.5*	O(3)-C(8)-N(2)	123.6(2)
C(4)-C(2)-H(2C)	*109.5*	O(3)-C(8)-C(6)	119.4(2)
C(4)-C(2)-H(2B)	*109.5*	N(2)-C(8)-C(6)	116.9(2)
H(2C)-C(2)-H(2B)	*109.5*	N(2)-C(9)-C(14)	113.25(18)
C(4)-C(2)-H(2A)	*109.5*	N(2)-C(9)-C(10)	109.7(2)
H(2C)-C(2)-H(2A)	*109.5*	C(14)-C(9)-C(10)	111.28(19)
H(2B)-C(2)-H(2A)	*109.5*	N(2)-C(9)-H(9)	107.4
C(4)-C(3)-H(3A)	*109.5*	C(14)-C(9)-H(9)	107.4
C(4)-C(3)-H(3B)	*109.5*	C(10)-C(9)-H(9)	107.4
H(3A)-C(3)-H(3B)	*109.5*	C(11)-C(10)-C(9)	111.9(2)
C(4)-C(3)-H(3C)	*109.5*	C(11)-C(10)-H(10B)	109.2
H(3A)-C(3)-H(3C)	*109.5*	C(9)-C(10)-H(10B)	109.2
H(3B)-C(3)-H(3C)	*109.5*	C(11)-C(10)-H(10A)	109.2
O(1)-C(4)-C(3)	111.1(2)	C(9)-C(10)-H(10A)	109.2
O(1)-C(4)-C(1)	108.8(2)	H(10B)-C(10)-H(10A)	107.9

C(12)-C(11)-C(10) 110.6(2)
C(12)-C(11)-H(11B) 109.5
C(10)-C(11)-H(11B) 109.5
C(12)-C(11)-H(11A) 109.5
C(10)-C(11)-H(11A) 109.5
H(11B)-C(11)-H(11A) 108.1
C(11)-C(12)-C(13) 110.6(2)
C(11)-C(12)-H(12A) 109.5
C(13)-C(12)-H(12A) 109.5
C(11)-C(12)-H(12B) 109.5
C(13)-C(12)-H(12B) 109.5
H(12A)-C(12)-H(12B) 108.1
C(12)-C(13)-C(14) 110.7(2)
C(12)-C(13)-H(13B) 109.5
C(14)-C(13)-H(13B) 109.5
C(12)-C(13)-H(13A) 109.5
C(14)-C(13)-H(13A) 109.5
H(13B)-C(13)-H(13A) 108.1
C(9)-C(14)-C(13) 108.8(2)
C(9)-C(14)-C(15) 111.09(18)
C(13)-C(14)-C(15) 115.1(2)
C(9)-C(14)-H(14) 107.2
C(13)-C(14)-H(14) 107.2
C(15)-C(14)-H(14) 107.2
C(18)-C(15)-C(16) 109.82(18)
C(18)-C(15)-C(14) 107.24(17)
C(16)-C(15)-C(14) 116.33(19)
C(18)-C(15)-H(15) 107.7
C(16)-C(15)-H(15) 107.7
C(14)-C(15)-H(15) 107.7
C(17)-C(16)-C(15) 112.0(2)
C(17)-C(16)-H(16A) 109.2
C(15)-C(16)-H(16A) 109.2
C(17)-C(16)-H(16B) 109.2
C(15)-C(16)-H(16B) 109.2
H(16A)-C(16)-H(16B) 107.9
C(16)-C(17)-H(17B) 109.5
C(16)-C(17)-H(17C) 109.5
H(17B)-C(17)-H(17C) 109.5
C(16)-C(17)-H(17A) 109.5
H(17B)-C(17)-H(17A) 109.5
H(17C)-C(17)-H(17A) 109.5
O(4)-C(18)-N(3) 122.6(2)
O(4)-C(18)-C(15) 122.2(2)
N(3)-C(18)-C(15) 115.08(18)
N(3)-C(19)-C(20) 109.24(19)
N(3)-C(19)-C(21) 113.83(18)
C(20)-C(19)-C(21) 109.12(18)
N(3)-C(19)-H(19) 108.2
C(20)-C(19)-H(19) 108.2
C(21)-C(19)-H(19) 108.2
C(19)-C(20)-H(20B) 109.5
C(19)-C(20)-H(20C) 109.5
H(20B)-C(20)-H(20C) 109.5
C(19)-C(20)-H(20A) 109.5
H(20B)-C(20)-H(20A) 109.5
H(20C)-C(20)-H(20A) 109.5
O(5)-C(21)-N(4) 124.0(2)
O(5)-C(21)-C(19) 118.12(19)
N(4)-C(21)-C(19) 117.66(18)
N(4)-C(22)-C(23) 109.84(17)
N(4)-C(22)-C(27) 111.25(18)
C(23)-C(22)-C(27) 110.80(18)
N(4)-C(22)-H(22) 108.3
C(23)-C(22)-H(22) 108.3
C(27)-C(22)-H(22) 108.3
C(22)-C(23)-C(24) 111.64(18)
C(22)-C(23)-H(23A) 109.3
C(24)-C(23)-H(23A) 109.3
C(22)-C(23)-H(23B) 109.3
C(24)-C(23)-H(23B) 109.3
H(23A)-C(23)-H(23B) 108.0
C(25)-C(24)-C(23) 110.70(19)
C(25)-C(24)-H(24B) 109.5
C(23)-C(24)-H(24B) 109.5
C(25)-C(24)-H(24A) 109.5
C(23)-C(24)-H(24A) 109.5
H(24B)-C(24)-H(24A) 108.1
C(26)-C(25)-C(24) 110.80(19)
C(26)-C(25)-H(25B) 109.5
C(24)-C(25)-H(25B) 109.5
C(26)-C(25)-H(25A) 109.5
C(24)-C(25)-H(25A) 109.5
H(25B)-C(25)-H(25A) 108.1
C(25)-C(26)-C(27) 111.73(18)
C(25)-C(26)-H(26A) 109.3
C(27)-C(26)-H(26A) 109.3
C(25)-C(26)-H(26B) 109.3
C(27)-C(26)-H(26B) 109.3
H(26A)-C(26)-H(26B) 107.9
C(26)-C(27)-C(22) 109.44(18)
C(26)-C(27)-C(28) 112.55(17)

C(22)-C(27)-C(28)	112.47(18)	O(7)-C(32)-C(33)	106.69(18)
C(26)-C(27)-H(27)	107.4	O(7)-C(32)-H(32A)	110.4
C(22)-C(27)-H(27)	107.4	C(33)-C(32)-H(32A)	110.4
C(28)-C(27)-H(27)	107.4	O(7)-C(32)-H(32B)	110.4
C(31)-C(28)-C(29)	111.56(18)	C(33)-C(32)-H(32B)	110.4
C(31)-C(28)-C(27)	108.53(18)	H(32A)-C(32)-H(32B)	108.6
C(29)-C(28)-C(27)	115.44(19)	C(34)-C(33)-C(38)	119.1(3)
C(31)-C(28)-H(28)	107.0	C(34)-C(33)-C(32)	121.3(2)
C(29)-C(28)-H(28)	107.0	C(38)-C(33)-C(32)	119.6(2)
C(27)-C(28)-H(28)	107.0	C(33)-C(34)-C(35)	120.9(3)
C(30)-C(29)-C(28)	111.9(2)	C(33)-C(34)-H(34)	119.6
C(30)-C(29)-H(29B)	109.2	C(35)-C(34)-H(34)	119.6
C(28)-C(29)-H(29B)	109.2	C(36)-C(35)-C(34)	119.0(3)
C(30)-C(29)-H(29A)	109.2	C(36)-C(35)-H(35)	120.5
C(28)-C(29)-H(29A)	109.2	C(34)-C(35)-H(35)	120.5
H(29B)-C(29)-H(29A)	107.9	C(35)-C(36)-C(37)	121.4(3)
C(29)-C(30)-H(30B)	109.5	C(35)-C(36)-Br(1)	119.2(2)
C(29)-C(30)-H(30C)	109.5	C(37)-C(36)-Br(1)	119.4(2)
H(30B)-C(30)-H(30C)	109.5	C(36)-C(37)-C(38)	119.5(3)
C(29)-C(30)-H(30A)	109.5	C(36)-C(37)-H(37)	120.3
H(30B)-C(30)-H(30A)	109.5	C(38)-C(37)-H(37)	120.3
H(30C)-C(30)-H(30A)	109.5	C(33)-C(38)-C(37)	120.1(3)
O(6)-C(31)-O(7)	123.2(2)	C(33)-C(38)-H(38)	120.0
O(6)-C(31)-C(28)	126.6(2)	C(37)-C(38)-H(38)	120.0
O(7)-C(31)-C(28)	110.26(19)		

Symmetry transformations used to generate equivalent atoms:

Table S32. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for gellman126. The anisotropic displacement factor exponent takes the form: $-2\pi^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Br(1)	50(1)	47(1)	81(1)	29(1)	-14(1)	11(1)
O(1)	36(1)	32(1)	23(1)	-2(1)	2(1)	21(1)
O(2)	37(1)	33(1)	30(1)	-2(1)	0(1)	23(1)
O(3)	40(1)	32(1)	32(1)	-7(1)	-13(1)	24(1)
O(4)	34(1)	27(1)	34(1)	2(1)	-1(1)	18(1)
O(5)	32(1)	29(1)	34(1)	-10(1)	-9(1)	18(1)
O(6)	24(1)	45(1)	35(1)	0(1)	-3(1)	13(1)
O(7)	25(1)	32(1)	35(1)	4(1)	0(1)	10(1)
N(1)	34(1)	29(1)	27(1)	-1(1)	1(1)	19(1)
N(2)	32(1)	28(1)	23(1)	-1(1)	-4(1)	20(1)
N(3)	27(1)	24(1)	24(1)	-4(1)	-4(1)	15(1)
N(4)	21(1)	24(1)	27(1)	-3(1)	-4(1)	12(1)
C(1)	67(2)	61(2)	37(2)	5(1)	19(1)	44(2)
C(2)	69(2)	47(2)	27(1)	-5(1)	-5(1)	39(1)
C(3)	58(2)	39(1)	41(2)	-8(1)	-17(1)	27(1)
C(4)	44(1)	36(1)	24(1)	-5(1)	1(1)	25(1)
C(5)	24(1)	28(1)	29(1)	0(1)	3(1)	12(1)
C(6)	32(1)	31(1)	28(1)	-6(1)	-5(1)	16(1)
C(7)	42(1)	34(1)	38(2)	-10(1)	-9(1)	8(1)
C(8)	33(1)	26(1)	25(1)	-8(1)	-5(1)	18(1)
C(9)	34(1)	39(1)	23(1)	3(1)	1(1)	25(1)
C(10)	66(2)	56(2)	21(1)	2(1)	3(1)	48(2)
C(11)	71(2)	93(2)	24(1)	15(1)	13(1)	64(2)
C(12)	50(2)	73(2)	30(1)	18(1)	11(1)	37(2)
C(13)	36(1)	48(1)	34(1)	14(1)	3(1)	22(1)
C(14)	28(1)	37(1)	25(1)	2(1)	-5(1)	20(1)
C(15)	26(1)	26(1)	25(1)	2(1)	-2(1)	14(1)
C(16)	30(1)	34(1)	41(2)	-3(1)	-10(1)	18(1)
C(17)	44(2)	44(2)	42(2)	-14(1)	-17(1)	19(1)
C(18)	26(1)	25(1)	24(1)	-3(1)	-4(1)	14(1)
C(19)	28(1)	30(1)	27(1)	-9(1)	-5(1)	17(1)
C(20)	35(1)	53(2)	29(1)	-1(1)	3(1)	27(1)
C(21)	20(1)	24(1)	29(1)	-4(1)	-2(1)	11(1)
C(22)	23(1)	21(1)	29(1)	0(1)	-2(1)	12(1)
C(23)	25(1)	25(1)	30(1)	1(1)	-1(1)	10(1)
C(24)	34(1)	36(1)	26(1)	4(1)	6(1)	20(1)
C(25)	32(1)	37(1)	25(1)	-3(1)	-3(1)	21(1)
C(26)	24(1)	34(1)	27(1)	-4(1)	-4(1)	15(1)
C(27)	23(1)	24(1)	24(1)	-2(1)	0(1)	12(1)
C(28)	24(1)	33(1)	23(1)	-4(1)	-2(1)	15(1)
C(29)	28(1)	39(1)	33(1)	-3(1)	-1(1)	20(1)

C(30)	41(1)	52(2)	40(2)	-7(1)	2(1)	31(1)
C(31)	26(1)	38(1)	23(1)	-2(1)	3(1)	16(1)
C(32)	26(1)	32(1)	36(1)	-1(1)	-1(1)	8(1)
C(33)	24(1)	34(1)	33(1)	6(1)	4(1)	7(1)
C(34)	31(1)	36(1)	44(2)	-2(1)	-2(1)	9(1)
C(35)	34(1)	35(1)	61(2)	8(1)	-1(1)	11(1)
C(36)	32(1)	44(2)	46(2)	23(1)	1(1)	10(1)
C(37)	33(1)	37(1)	35(1)	7(1)	2(1)	6(1)
C(38)	32(1)	30(1)	34(1)	4(1)	4(1)	7(1)

Table S33. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for gellman126.

	x	y	z	U(eq)	
H(1)	6280	6286	6214	34	
H(2)	7141	7204	5171	31	
H(3)	6542	8188	5924	29	
H(4)	6050	8122	3725	28	
H(1C)	6109	7038	10638	74	
H(1A)	5755	6473	9749	74	
H(1B)	5889	7131	9345	74	
H(2C)	6942	6793	10776	65	
H(2B)	7300	6797	9588	65	
H(2A)	6626	6252	9821	65	
H(3A)	6925	7925	8828	67	
H(3B)	7455	7767	8687	67	
H(3C)	7253	7859	10000	67	
H(6)	5693	6828	5247	36	
H(7B)	5584	6241	3511	64	
H(7A)	5392	5826	4694	64	
H(7C)	6036	6043	4079	64	
H(9)	7977	7894	4346	34	
H(10B)		7606	7243	2669	48
H(10A)		7242	7567	2215	48
H(11B)		8537	8122	2290	63
H(11A)		8137	7951	1086	63
H(12A)		7862	8684	1519	57
H(12B)		8586	9002	1502	57
H(13B)		8276	9330	3220	47
H(13A)		8600	8961	3643	47
H(14)	7309	8441	3468	33	
H(15)	7435	8309	5752	30	
H(16A)		8517	9015	5732	41

H(16B)	8372	9540	5394	41
H(17B)	7823	9351	7241	67
H(17C)	8519	9557	7478	67
H(17A)	8011	8858	7584	67
H(19)	6333	9127	6024	33
H(20B)	5304	8370	6374	55
H(20C)	5764	8303	7254	55
H(20A)	5483	7862	6118	55
H(22)	5574	8663	2244	29
H(23A)	6065	7913	1539	33
H(23B)	6411	8635	1417	33
H(24B)	5711	8575	-48	37
H(24A)	5984	8160	-476	37
H(25B)	4904	7640	-704	35
H(25A)	5110	7279	184	35
H(26A)	4287	7320	1054	34
H(26B)	4644	8043	950	34
H(27)	5014	7369	2450	28
H(28)	4809	7863	4120	32
H(29B)	4631	8552	2864	38
H(29A)	3971	7986	2610	38
H(30B)	3922	8557	4232	61
H(30C)	4417	8473	4957	61
H(30A)	3760	7902	4713	61
H(32A)	3679	5920	3815	41
H(32B)	3452	6144	4953	41
H(34)	3827	5125	4613	49
H(35)	4290	4775	5960	56
H(37)	5009	6357	7762	48
H(38)	4506	6690	6475	43

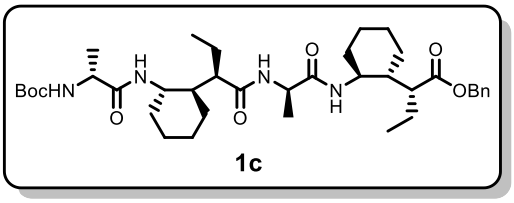
Table S34. Torsion angles [°] for gellman126.

C(5)-O(1)-C(4)-C(3)	-57.5(3)	C(8)-N(2)-C(9)-C(14)	56.9(3)
C(5)-O(1)-C(4)-C(1)	67.5(3)	C(8)-N(2)-C(9)-C(10)	-68.1(3)
C(5)-O(1)-C(4)-C(2)	-175.6(2)	N(2)-C(9)-C(10)-C(11)	-178.70(19)
C(6)-N(1)-C(5)-O(2)	-0.2(3)	C(14)-C(9)-C(10)-C(11)	55.2(3)
C(6)-N(1)-C(5)-O(1)	-179.74(19)	C(9)-C(10)-C(11)-C(12)	-54.9(3)
C(4)-O(1)-C(5)-O(2)	5.3(3)	C(10)-C(11)-C(12)-C(13)	57.1(3)
C(4)-O(1)-C(5)-N(1)	-175.10(19)	C(11)-C(12)-C(13)-C(14)	-59.9(3)
C(5)-N(1)-C(6)-C(8)	76.5(3)	N(2)-C(9)-C(14)-C(13)	179.71(19)
C(5)-N(1)-C(6)-C(7)	-162.0(2)	C(10)-C(9)-C(14)-C(13)	-56.1(3)
C(9)-N(2)-C(8)-O(3)	-1.0(3)	N(2)-C(9)-C(14)-C(15)	52.0(2)
C(9)-N(2)-C(8)-C(6)	175.9(2)	C(10)-C(9)-C(14)-C(15)	176.2(2)
N(1)-C(6)-C(8)-O(3)	-162.98(19)	C(12)-C(13)-C(14)-C(9)	58.7(3)
C(7)-C(6)-C(8)-O(3)	75.0(3)	C(12)-C(13)-C(14)-C(15)	-175.9(2)
N(1)-C(6)-C(8)-N(2)	19.9(3)	C(9)-C(14)-C(15)-C(18)	-133.55(18)
C(7)-C(6)-C(8)-N(2)	-102.1(2)	C(13)-C(14)-C(15)-C(18)	102.3(2)

C(9)-C(14)-C(15)-C(16)	103.1(2)
C(13)-C(14)-C(15)-C(16)	-21.1(3)
C(18)-C(15)-C(16)-C(17)	67.1(3)
C(14)-C(15)-C(16)-C(17)	-170.9(2)
C(19)-N(3)-C(18)-O(4)	-2.8(3)
C(19)-N(3)-C(18)-C(15)	-179.59(19)
C(16)-C(15)-C(18)-O(4)	54.1(3)
C(14)-C(15)-C(18)-O(4)	-73.2(3)
C(16)-C(15)-C(18)-N(3)	-129.1(2)
C(14)-C(15)-C(18)-N(3)	103.7(2)
C(18)-N(3)-C(19)-C(20)	-168.5(2)
C(18)-N(3)-C(19)-C(21)	69.3(3)
C(22)-N(4)-C(21)-O(5)	-1.9(3)
C(22)-N(4)-C(21)-C(19)	172.50(18)
N(3)-C(19)-C(21)-O(5)	-151.5(2)
C(20)-C(19)-C(21)-O(5)	86.2(2)
N(3)-C(19)-C(21)-N(4)	33.7(3)
C(20)-C(19)-C(21)-N(4)	-88.6(2)
C(21)-N(4)-C(22)-C(23)	135.3(2)
C(21)-N(4)-C(22)-C(27)	-101.7(2)
N(4)-C(22)-C(23)-C(24)	-179.92(17)
C(27)-C(22)-C(23)-C(24)	56.8(2)
C(22)-C(23)-C(24)-C(25)	-55.7(2)
C(23)-C(24)-C(25)-C(26)	55.2(3)
C(24)-C(25)-C(26)-C(27)	-57.0(3)
C(25)-C(26)-C(27)-C(22)	57.3(2)
C(25)-C(26)-C(27)-C(28)	-176.89(19)
N(4)-C(22)-C(27)-C(26)	-179.24(17)
C(23)-C(22)-C(27)-C(26)	-56.7(2)
N(4)-C(22)-C(27)-C(28)	54.9(2)
C(23)-C(22)-C(27)-C(28)	177.38(18)
C(26)-C(27)-C(28)-C(31)	78.2(2)
C(22)-C(27)-C(28)-C(31)	-157.63(18)
C(26)-C(27)-C(28)-C(29)	-47.9(3)
C(22)-C(27)-C(28)-C(29)	76.3(2)
C(31)-C(28)-C(29)-C(30)	73.4(3)
C(27)-C(28)-C(29)-C(30)	-162.1(2)
C(32)-O(7)-C(31)-O(6)	-2.2(3)
C(32)-O(7)-C(31)-C(28)	179.09(19)
C(29)-C(28)-C(31)-O(6)	31.5(3)
C(27)-C(28)-C(31)-O(6)	-96.8(3)
C(29)-C(28)-C(31)-O(7)	-149.9(2)
C(27)-C(28)-C(31)-O(7)	81.8(2)
C(31)-O(7)-C(32)-C(33)	-176.8(2)
O(7)-C(32)-C(33)-C(34)	-136.3(2)
O(7)-C(32)-C(33)-C(38)	44.5(3)
C(38)-C(33)-C(34)-C(35)	0.1(4)
C(32)-C(33)-C(34)-C(35)	-179.1(2)
C(33)-C(34)-C(35)-C(36)	-2.0(4)
C(34)-C(35)-C(36)-C(37)	2.2(4)
C(34)-C(35)-C(36)-Br(1)	-177.3(2)
C(35)-C(36)-C(37)-C(38)	-0.5(4)
Br(1)-C(36)-C(37)-C(38)	178.9(2)
C(34)-C(33)-C(38)-C(37)	1.6(4)
C(32)-C(33)-C(38)-C(37)	-179.2(2)
C(36)-C(37)-C(38)-C(33)	-1.5(4)

Symmetry transformations used to generate equivalent atoms:

Crystallographic Experimental Section for Compound 1c



1c

Data Collection

A colorless crystal with approximate dimensions 0.43 x 0.28 x 0.11 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker SMART APEXII diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 41 frames collected at intervals of 0.6º in a 25º range about ω with the exposure time of 10 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9700 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.82 Å. A total of 125656 data were harvested by collecting 19 sets of frames with 0.5º scans in ω with an exposure time 30-60 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were uniquely consistent for the space group $P2_12_12_1$ that yielded chemically reasonable and computationally stable results of refinement [2,3].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms were refined with anisotropic displacement coefficients. All hydrogen atoms were included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

There is also one molecule of solvate water per two molecules of the foldamer in the asymmetric unit. The water molecule was refined with an idealized geometry.

One phenyl group is disordered over two positions with the major component being occupied 62.3(4)% of the time. Restraints and constraints were used for the refinement of the disordered phenyl groups.

The final least-squares refinement of 911 parameters against 15507 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0405 and 0.1000, respectively. The final difference Fourier map was featureless.

The molecular diagram is drawn with 40% probability ellipsoids.

References

[1] Bruker-AXS. (2007) APEX2, SADABS, and SAINT Software Reference Manuals. Bruker-AXS, Madison, Wisconsin, USA.

[2] Sheldrick, G. M. (2008) SHELXL. *Acta Cryst.* **A64**, 112-122.

[3] Dolomanov, O.V.; Bourhis, L.J.; Gildea, R.J.; Howard, J.A.K.; Puschmann, H. "OLEX2: a complete structure solution, refinement and analysis program". *J. Appl. Cryst.* (2009) **42**, *339-341*.

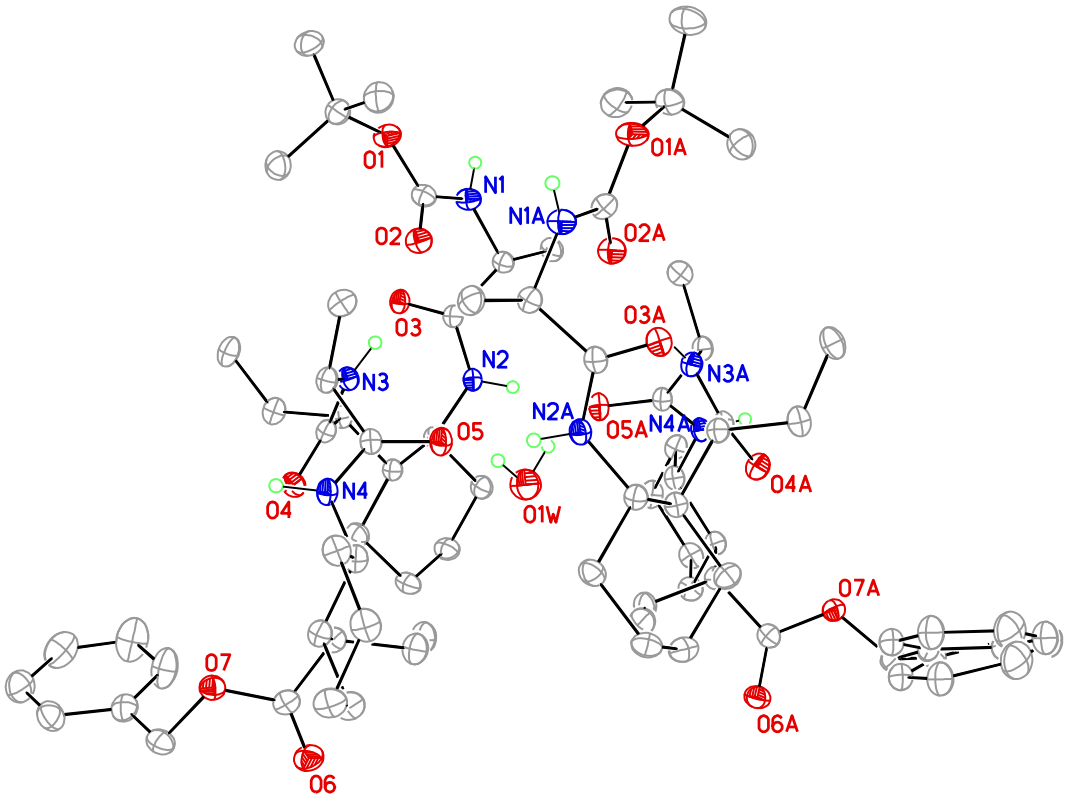


Figure S22. A molecular drawing of the unit cell content (note the water molecule and disorder). All H atoms connected to C atoms are omitted. The molecular diagram is drawn with 30% probability ellipsoids.

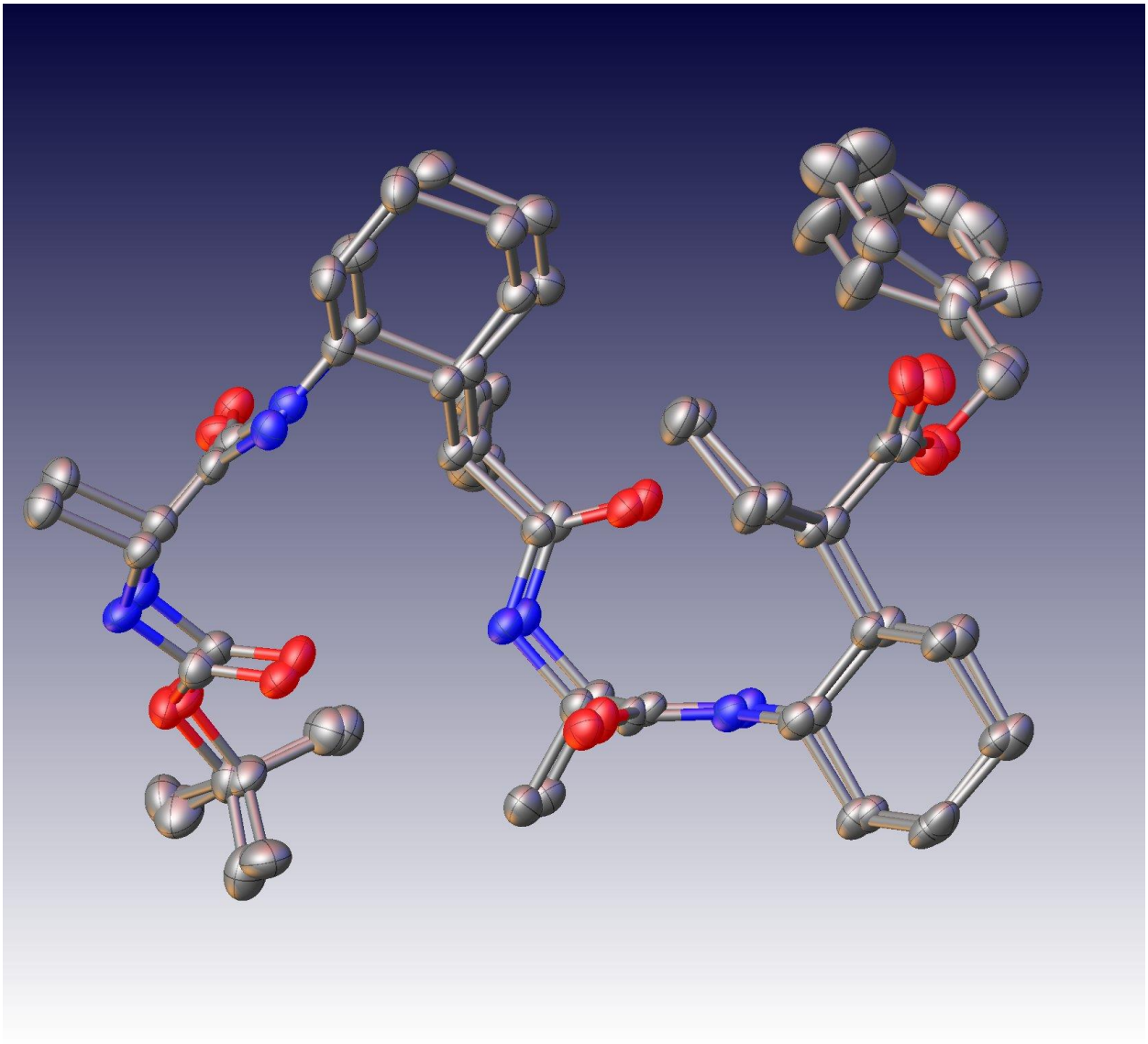


Figure S23. A molecular drawing of the two foldamer molecules overlapped. All H atoms and connected to C atoms are omitted. The molecular diagram is drawn with 50% probability ellipsoids.

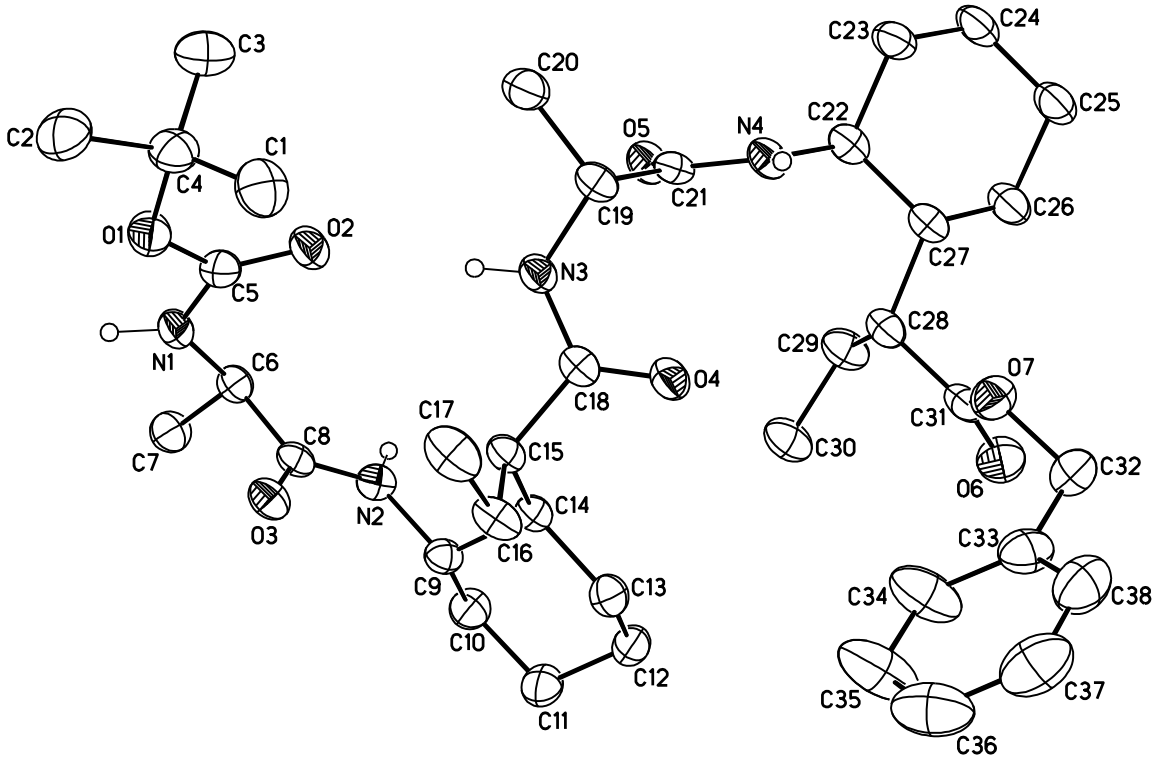


Figure S24. A molecular drawing of the first foldamer molecule. All H atoms connected to C atoms are omitted. The molecular diagram is drawn with 50% probability ellipsoids.

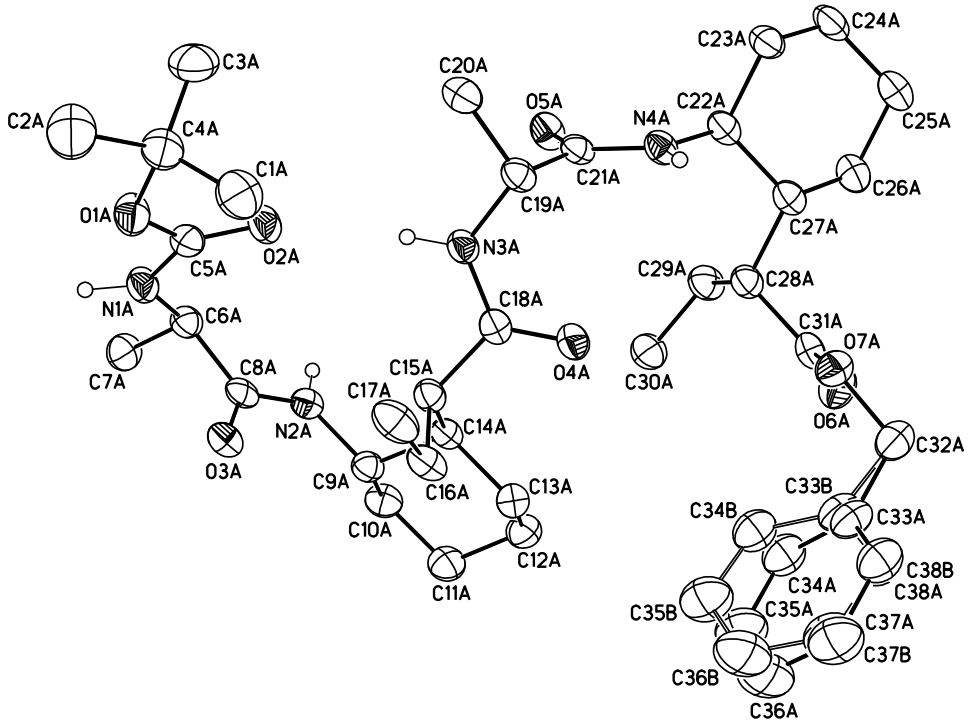


Figure S25. A molecular drawing of the second foldamer molecule. All H atoms connected to C atoms and the minor components of the disordered atoms are omitted. The molecular diagram is drawn with 50% probability ellipsoids.

Table S35. Crystal data and structure refinement for gellman124a.

Identification code	**gellman124a**	
Empirical formula	$C_{38}\ H_{60}\ N_4\ O_7$. 1/2 H_2O	
Formula weight	693.91	
Temperature	100(1) K	
Wavelength	1.54178 Å	
Crystal system	Orthorhombic	
Space group	$P2_12_12_1$	
Unit cell dimensions	a = 16.5265(5) Å	α= 90°.
	b = 17.3446(5) Å	β= 90°.
	c = 28.1866(9) Å	γ = 90°.
Volume	8079.6(4) $Å^3$	
Z	8	
Density (calculated)	1.141 Mg/m^3	
Absorption coefficient	0.636 mm^{-1}	
F(000)	3016	
Crystal size	0.43 x 0.28 x 0.11 mm^3	
Theta range for data collection	2.99 to 71.68°.	
Index ranges	-17<=h<=19, -21<=k<=21, -34<=l<=33	
Reflections collected	125656	
Independent reflections	15507 [R(int) = 0.0523]	
Completeness to theta = 67.00°	99.5 %	
Absorption correction	Empirical with SADABS	
Max. and min. transmission	0.9316 and 0.7714	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	15507 / 1 / 911	
Goodness-of-fit on F^2	1.039	
Final R indices [I>2sigma(I)]	R1 = 0.0405, wR2 = 0.0917	
R indices (all data)	R1 = 0.0540, wR2 = 0.1000	
Absolute structure parameter (Flack x)	-0.06(11)	
Absolute structure parameter (Hooft y)	-0.09(04)	
Largest diff. peak and hole	0.211 and -0.185 $e.Å^{-3}$	

Table S36. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for gellman124a. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
O(1)	-780(1)	-9264(1)	-1540(1)	38(1)
O(2)	-1355(1)	-9658(1)	-843(1)	36(1)
O(3)	-210(1)	-8247(1)	-336(1)	31(1)
O(4)	-379(1)	-11035(1)	689(1)	32(1)
O(5)	-2558(1)	-11469(1)	326(1)	34(1)
O(6)	-946(1)	-11926(1)	2371(1)	44(1)
O(7)	-208(1)	-12582(1)	1837(1)	36(1)
N(1)	-1314(1)	-8395(1)	-1061(1)	32(1)
N(2)	-1205(1)	-8329(1)	212(1)	28(1)
N(3)	-1021(1)	-10797(1)	-2(1)	28(1)
N(4)	-1718(1)	-12433(1)	559(1)	28(1)
C(1)	-73(2)	-10486(1)	-1398(1)	46(1)
C(2)	-182(2)	-9862(1)	-2190(1)	48(1)
C(3)	-1414(2)	-10451(1)	-1820(1)	50(1)
C(4)	-620(1)	-10040(1)	-1729(1)	38(1)
C(5)	-1160(1)	-9158(1)	-1124(1)	32(1)
C(6)	-1586(1)	-8121(1)	-605(1)	29(1)
C(7)	-1793(1)	-7261(1)	-642(1)	36(1)
C(8)	-935(1)	-8243(1)	-231(1)	27(1)
C(9)	-678(1)	-8420(1)	624(1)	28(1)
C(10)	-1024(1)	-7929(1)	1027(1)	35(1)
C(11)	-535(2)	-8001(1)	1482(1)	39(1)
C(12)	-466(2)	-8845(1)	1629(1)	38(1)
C(13)	-101(1)	-9325(1)	1229(1)	34(1)
C(14)	-599(1)	-9272(1)	769(1)	27(1)
C(15)	-254(1)	-9758(1)	355(1)	28(1)
C(16)	671(1)	-9767(1)	325(1)	35(1)
C(17)	972(1)	-10172(1)	-122(1)	43(1)
C(18)	-559(1)	-10586(1)	371(1)	27(1)
C(19)	-1263(1)	-11597(1)	-64(1)	29(1)
C(20)	-1588(1)	-11719(1)	-565(1)	38(1)
C(21)	-1908(1)	-11824(1)	300(1)	26(1)
C(22)	-2233(1)	-12765(1)	925(1)	31(1)
C(23)	-2520(1)	-13565(1)	771(1)	35(1)
C(24)	-3043(1)	-13945(1)	1150(1)	39(1)
C(25)	-2606(2)	-13983(1)	1623(1)	40(1)
C(26)	-2318(1)	-13187(1)	1775(1)	38(1)
C(27)	-1774(1)	-12818(1)	1398(1)	30(1)
C(28)	-1426(1)	-12029(1)	1560(1)	31(1)
C(29)	-2066(1)	-11421(1)	1683(1)	41(1)
C(30)	-1697(2)	-10615(1)	1735(1)	44(1)
C(31)	-857(1)	-12154(1)	1971(1)	32(1)
C(32)	409(1)	-12736(1)	2194(1)	41(1)
C(33)	1173(2)	-12324(1)	2066(1)	41(1)
C(34)	1157(2)	-11568(1)	1909(1)	62(1)
C(35)	1864(2)	-11197(2)	1790(1)	77(1)
C(36)	2597(2)	-11575(2)	1834(1)	68(1)
C(37)	2614(2)	-12319(2)	1979(1)	62(1)
C(38)	1909(2)	-12691(2)	2095(1)	52(1)
O(1A)	-4557(1)	-10615(1)	-1535(1)	39(1)
O(2A)	-4096(1)	-10286(1)	-798(1)	35(1)

O(3A)	-5297(1)	-11748(1)	-338(1)	31(1)
O(4A)	-5109(1)	-8939(1)	704(1)	34(1)
O(5A)	-2938(1)	-8465(1)	374(1)	34(1)
O(6A)	-4612(1)	-8060(1)	2408(1)	45(1)
O(7A)	-5380(1)	-7506(1)	1848(1)	35(1)
N(1A)	-4155(1)	-11531(1)	-1046(1)	32(1)
N(2A)	-4324(1)	-11665(1)	223(1)	28(1)
N(3A)	-4445(1)	-9131(1)	17(1)	28(1)
N(4A)	-3776(1)	-7505(1)	614(1)	28(1)
C(1A)	-5255(2)	-9386(1)	-1419(1)	46(1)
C(2A)	-4936(2)	-9935(1)	-2212(1)	56(1)
C(3A)	-3811(2)	-9429(1)	-1711(1)	49(1)
C(4A)	-4631(2)	-9816(1)	-1711(1)	38(1)
C(5A)	-4265(1)	-10762(1)	-1100(1)	31(1)
C(6A)	-3909(1)	-11842(1)	-593(1)	30(1)
C(7A)	-3710(1)	-12699(1)	-648(1)	37(1)
C(8A)	-4577(1)	-11742(1)	-226(1)	27(1)
C(9A)	-4868(1)	-11554(1)	625(1)	29(1)
C(10A)	-4584(1)	-12059(1)	1036(1)	36(1)
C(11A)	-5110(2)	-11957(1)	1476(1)	39(1)
C(12A)	-5121(1)	-11108(1)	1623(1)	39(1)
C(13A)	-5432(1)	-10609(1)	1216(1)	35(1)
C(14A)	-4914(1)	-10696(1)	767(1)	28(1)
C(15A)	-5210(1)	-10200(1)	343(1)	28(1)
C(16A)	-6133(1)	-10184(1)	275(1)	34(1)
C(17A)	-6378(1)	-9778(1)	-183(1)	40(1)
C(18A)	-4912(1)	-9373(1)	378(1)	27(1)
C(19A)	-4220(1)	-8325(1)	-25(1)	29(1)
C(20A)	-3900(1)	-8158(1)	-522(1)	35(1)
C(21A)	-3583(1)	-8108(1)	347(1)	26(1)
C(22A)	-3272(1)	-7205(1)	996(1)	30(1)
C(23A)	-2937(1)	-6413(1)	858(1)	35(1)
C(24A)	-2416(1)	-6076(1)	1254(1)	39(1)
C(25A)	-2898(1)	-6019(1)	1712(1)	39(1)
C(26A)	-3233(1)	-6802(1)	1853(1)	38(1)
C(27A)	-3754(1)	-7159(1)	1461(1)	31(1)
C(28A)	-4099(1)	-7958(1)	1602(1)	31(1)
C(29A)	-3461(1)	-8561(1)	1735(1)	37(1)
C(30A)	-3812(2)	-9374(1)	1724(1)	40(1)
C(31A)	-4701(1)	-7860(1)	2004(1)	31(1)
C(32A)	-6042(1)	-7406(1)	2184(1)	39(1)
C(33A)	-6750(1)	-7917(1)	2072(1)	37(1)
C(34A)	-6622(1)	-8708(2)	2093(1)	43(1)
C(35A)	-7257(2)	-9214(1)	2007(1)	56(1)
C(36A)	-8021(1)	-8929(2)	1899(1)	66(1)
C(37A)	-8150(1)	-8138(2)	1877(1)	56(1)
C(38A)	-7514(2)	-7632(1)	1963(2)	43(1)
C(33B)	-6687(1)	-7962(2)	2012(2)	37(1)
C(34B)	-6528(1)	-8711(2)	1864(1)	43(1)
C(35B)	-7158(1)	-9183(2)	1714(2)	56(1)
C(36B)	-7947(1)	-8905(3)	1711(2)	66(1)
C(37B)	-8106(1)	-8156(3)	1859(2)	56(1)
C(38B)	-7476(2)	-7684(2)	2010(2)	43(1)
O(1W)	-2746(1)	-9987(1)	755(1)	56(1)

Table S37. Bond lengths [Å] and angles [°] for gellman124a.

O(1)-C(5)	1.344(3)	C(14)-C(15)	1.547(3)
O(1)-C(4)	1.472(2)	C(14)-H(14A)	1.0000
O(2)-C(5)	1.219(2)	C(15)-C(18)	1.523(2)
O(3)-C(8)	1.233(2)	C(15)-C(16)	1.531(3)
O(4)-C(18)	1.223(2)	C(15)-H(15A)	1.0000
O(5)-C(21)	1.239(2)	C(16)-C(17)	1.525(3)
O(6)-C(31)	1.202(2)	C(16)-H(16C)	0.9900
O(7)-C(31)	1.359(3)	C(16)-H(16D)	0.9900
O(7)-C(32)	1.457(3)	C(17)-H(17D)	0.9800
N(1)-C(5)	1.357(3)	C(17)-H(17F)	0.9800
N(1)-C(6)	1.443(3)	C(17)-H(17E)	0.9800
N(1)-H(1)	0.82(2)	C(19)-C(20)	1.525(3)
N(2)-C(8)	1.335(3)	C(19)-C(21)	1.533(3)
N(2)-C(9)	1.460(2)	C(19)-H(19A)	1.0000
N(2)-H(2)	0.85(2)	C(20)-H(20D)	0.9800
N(3)-C(18)	1.350(3)	C(20)-H(20F)	0.9800
N(3)-C(19)	1.454(2)	C(20)-H(20E)	0.9800
N(3)-H(3)	0.83(2)	C(22)-C(23)	1.529(3)
N(4)-C(21)	1.321(2)	C(22)-C(27)	1.537(3)
N(4)-C(22)	1.457(3)	C(22)-H(22A)	1.0000
N(4)-H(4)	0.90(2)	C(23)-C(24)	1.523(3)
C(1)-C(4)	1.514(3)	C(23)-H(23C)	0.9900
C(1)-H(1B)	0.9800	C(23)-H(23D)	0.9900
C(1)-H(1D)	0.9800	C(24)-C(25)	1.519(3)
C(1)-H(1C)	0.9800	C(24)-H(24C)	0.9900
C(2)-C(4)	1.518(3)	C(24)-H(24D)	0.9900
C(2)-H(2C)	0.9800	C(25)-C(26)	1.522(3)
C(2)-H(2D)	0.9800	C(25)-H(25C)	0.9900
C(2)-H(2B)	0.9800	C(25)-H(25D)	0.9900
C(3)-C(4)	1.514(3)	C(26)-C(27)	1.532(3)
C(3)-H(3C)	0.9800	C(26)-H(26D)	0.9900
C(3)-H(3D)	0.9800	C(26)-H(26C)	0.9900
C(3)-H(3B)	0.9800	C(27)-C(28)	1.553(3)
C(6)-C(8)	1.522(3)	C(27)-H(27A)	1.0000
C(6)-C(7)	1.534(3)	C(28)-C(31)	1.508(3)
C(6)-H(6)	1.0000	C(28)-C(29)	1.534(3)
C(7)-H(7B)	0.9800	C(28)-H(28A)	1.0000
C(7)-H(7C)	0.9800	C(29)-C(30)	1.532(3)
C(7)-H(7A)	0.9800	C(29)-H(29C)	0.9900
C(9)-C(10)	1.530(3)	C(29)-H(29D)	0.9900
C(9)-C(14)	1.539(3)	C(30)-H(30D)	0.9800
C(9)-H(9)	1.0000	C(30)-H(30F)	0.9800
C(10)-C(11)	1.522(3)	C(30)-H(30E)	0.9800
C(10)-H(10C)	0.9900	C(32)-C(33)	1.495(3)
C(10)-H(10D)	0.9900	C(32)-H(32F)	0.9900
C(11)-C(12)	1.526(3)	C(32)-H(32E)	0.9900
C(11)-H(11D)	0.9900	C(33)-C(38)	1.375(3)
C(11)-H(11C)	0.9900	C(33)-C(34)	1.384(4)
C(12)-C(13)	1.528(3)	C(34)-C(35)	1.375(4)
C(12)-H(12D)	0.9900	C(34)-H(34B)	0.9500
C(12)-H(12C)	0.9900	C(35)-C(36)	1.383(4)
C(13)-C(14)	1.539(3)	C(35)-H(35B)	0.9500
C(13)-H(13D)	0.9900	C(36)-C(37)	1.355(4)
C(13)-H(13C)	0.9900	C(36)-H(36B)	0.9500

C(37)-C(38)	1.371(4)	C(13A)-H(13B)	0.9900
C(37)-H(37B)	0.9500	C(14A)-C(15A)	1.552(3)
C(38)-H(38B)	0.9500	C(14A)-H(14)	1.0000
O(1A)-C(5A)	1.343(2)	C(15A)-C(18A)	1.520(3)
O(1A)-C(4A)	1.475(2)	C(15A)-C(16A)	1.538(3)
O(2A)-C(5A)	1.218(2)	C(15A)-H(15)	1.0000
O(3A)-C(8A)	1.232(2)	C(16A)-C(17A)	1.524(3)
O(4A)-C(18A)	1.229(2)	C(16A)-H(16B)	0.9900
O(5A)-C(21A)	1.235(2)	C(16A)-H(16A)	0.9900
O(6A)-C(31A)	1.202(2)	C(17A)-H(17B)	0.9800
O(7A)-C(31A)	1.353(3)	C(17A)-H(17A)	0.9800
O(7A)-C(32A)	1.457(2)	C(17A)-H(17C)	0.9800
N(1A)-C(5A)	1.355(3)	C(19A)-C(20A)	1.525(3)
N(1A)-C(6A)	1.445(3)	C(19A)-C(21A)	1.532(3)
N(1A)-H(1A)	0.82(2)	C(19A)-H(19)	1.0000
N(2A)-C(8A)	1.338(3)	C(20A)-H(20A)	0.9800
N(2A)-C(9A)	1.460(2)	C(20A)-H(20C)	0.9800
N(2A)-H(2A)	0.83(2)	C(20A)-H(20B)	0.9800
N(3A)-C(18A)	1.346(3)	C(22A)-C(23A)	1.532(3)
N(3A)-C(19A)	1.452(2)	C(22A)-C(27A)	1.535(3)
N(3A)-H(3A)	0.84(2)	C(22A)-H(22)	1.0000
N(4A)-C(21A)	1.329(2)	C(23A)-C(24A)	1.525(3)
N(4A)-C(22A)	1.458(3)	C(23A)-H(23A)	0.9900
N(4A)-H(4A)	0.88(2)	C(23A)-H(23B)	0.9900
C(1A)-C(4A)	1.517(3)	C(24A)-C(25A)	1.521(3)
C(1A)-H(1AB)	0.9800	C(24A)-H(24B)	0.9900
C(1A)-H(1AC)	0.9800	C(24A)-H(24A)	0.9900
C(1A)-H(1AA)	0.9800	C(25A)-C(26A)	1.519(3)
C(2A)-C(4A)	1.513(3)	C(25A)-H(25B)	0.9900
C(2A)-H(2AA)	0.9800	C(25A)-H(25A)	0.9900
C(2A)-H(2AC)	0.9800	C(26A)-C(27A)	1.533(3)
C(2A)-H(2AB)	0.9800	C(26A)-H(26B)	0.9900
C(3A)-C(4A)	1.512(3)	C(26A)-H(26A)	0.9900
C(3A)-H(3AA)	0.9800	C(27A)-C(28A)	1.551(3)
C(3A)-H(3AC)	0.9800	C(27A)-H(27)	1.0000
C(3A)-H(3AB)	0.9800	C(28A)-C(31A)	1.515(3)
C(6A)-C(8A)	1.523(3)	C(28A)-C(29A)	1.531(3)
C(6A)-C(7A)	1.530(3)	C(28A)-H(28)	1.0000
C(6A)-H(6A)	1.0000	C(29A)-C(30A)	1.526(3)
C(7A)-H(7AC)	0.9800	C(29A)-H(29A)	0.9900
C(7A)-H(7AB)	0.9800	C(29A)-H(29B)	0.9900
C(7A)-H(7AA)	0.9800	C(30A)-H(30A)	0.9800
C(9A)-C(10A)	1.526(3)	C(30A)-H(30C)	0.9800
C(9A)-C(14A)	1.542(3)	C(30A)-H(30B)	0.9800
C(9A)-H(9A)	1.0000	C(32A)-C(33A)	1.502(3)
C(10A)-C(11A)	1.524(3)	C(32A)-C(33B)	1.517(4)
C(10A)-H(10B)	0.9900	C(32A)-H(32A)	0.9900
C(10A)-H(10A)	0.9900	C(32A)-H(32B)	0.9900
C(11A)-C(12A)	1.530(3)	C(32A)-H(32C)	0.9900
C(11A)-H(11A)	0.9900	C(32A)-H(32D)	0.9900
C(11A)-H(11B)	0.9900	C(33A)-C(34A)	1.3900
C(12A)-C(13A)	1.527(3)	C(33A)-C(38A)	1.3900
C(12A)-H(12A)	0.9900	C(34A)-C(35A)	1.3900
C(12A)-H(12B)	0.9900	C(34A)-H(34)	0.9500
C(13A)-C(14A)	1.535(3)	C(35A)-C(36A)	1.3900
C(13A)-H(13A)	0.9900	C(35A)-H(35)	0.9500

C(36A)-C(37A)	1.3900	C(35B)-C(36B)	1.3900
C(36A)-H(36)	0.9500	C(35B)-H(35A)	0.9500
C(37A)-C(38A)	1.3900	C(36B)-C(37B)	1.3900
C(37A)-H(37)	0.9500	C(36B)-H(36A)	0.9500
C(38A)-H(38)	0.9500	C(37B)-C(38B)	1.3900
C(33B)-C(34B)	1.3900	C(37B)-H(37A)	0.9500
C(33B)-C(38B)	1.3900	C(38B)-H(38A)	0.9500
C(34B)-C(35B)	1.3900	O(1W)-H(1W)	0.9584
C(34B)-H(34A)	0.9500	O(1W)-H(2W)	0.9584

C(5)-O(1)-C(4)	121.74(16)	C(8)-C(6)-H(6)	109.0
C(31)-O(7)-C(32)	117.34(16)	C(7)-C(6)-H(6)	109.0
C(5)-N(1)-C(6)	119.65(17)	C(6)-C(7)-H(7B)	109.5
C(5)-N(1)-H(1)	113.3(17)	C(6)-C(7)-H(7C)	109.5
C(6)-N(1)-H(1)	118.6(16)	H(7B)-C(7)-H(7C)	109.5
C(8)-N(2)-C(9)	123.80(17)	C(6)-C(7)-H(7A)	109.5
C(8)-N(2)-H(2)	119.7(15)	H(7B)-C(7)-H(7A)	109.5
C(9)-N(2)-H(2)	116.4(15)	H(7C)-C(7)-H(7A)	109.5
C(18)-N(3)-C(19)	120.56(17)	O(3)-C(8)-N(2)	123.26(18)
C(18)-N(3)-H(3)	122.7(16)	O(3)-C(8)-C(6)	121.40(17)
C(19)-N(3)-H(3)	116.6(16)	N(2)-C(8)-C(6)	115.34(17)
C(21)-N(4)-C(22)	124.64(17)	N(2)-C(9)-C(10)	107.91(16)
C(21)-N(4)-H(4)	118.3(14)	N(2)-C(9)-C(14)	111.35(15)
C(22)-N(4)-H(4)	117.0(14)	C(10)-C(9)-C(14)	111.66(16)
C(4)-C(1)-H(1B)	109.5	N(2)-C(9)-H(9)	108.6
C(4)-C(1)-H(1D)	109.5	C(10)-C(9)-H(9)	108.6
H(1B)-C(1)-H(1D)	109.5	C(14)-C(9)-H(9)	108.6
C(4)-C(1)-H(1C)	109.5	C(11)-C(10)-C(9)	112.46(17)
H(1B)-C(1)-H(1C)	109.5	C(11)-C(10)-H(10C)	109.1
H(1D)-C(1)-H(1C)	109.5	C(9)-C(10)-H(10C)	109.1
C(4)-C(2)-H(2C)	109.5	C(11)-C(10)-H(10D)	109.1
C(4)-C(2)-H(2D)	109.5	C(9)-C(10)-H(10D)	109.1
H(2C)-C(2)-H(2D)	109.5	H(10C)-C(10)-H(10D)	107.8
C(4)-C(2)-H(2B)	109.5	C(10)-C(11)-C(12)	110.31(17)
H(2C)-C(2)-H(2B)	109.5	C(10)-C(11)-H(11D)	109.6
H(2D)-C(2)-H(2B)	109.5	C(12)-C(11)-H(11D)	109.6
C(4)-C(3)-H(3C)	109.5	C(10)-C(11)-H(11C)	109.6
C(4)-C(3)-H(3D)	109.5	C(12)-C(11)-H(11C)	109.6
H(3C)-C(3)-H(3D)	109.5	H(11D)-C(11)-H(11C)	108.1
C(4)-C(3)-H(3B)	109.5	C(11)-C(12)-C(13)	110.59(17)
H(3C)-C(3)-H(3B)	109.5	C(11)-C(12)-H(12D)	109.5
H(3D)-C(3)-H(3B)	109.5	C(13)-C(12)-H(12D)	109.5
O(1)-C(4)-C(1)	110.57(17)	C(11)-C(12)-H(12C)	109.5
O(1)-C(4)-C(3)	109.72(18)	C(13)-C(12)-H(12C)	109.5
C(1)-C(4)-C(3)	112.4(2)	H(12D)-C(12)-H(12C)	108.1
O(1)-C(4)-C(2)	102.10(17)	C(12)-C(13)-C(14)	112.22(17)
C(1)-C(4)-C(2)	110.2(2)	C(12)-C(13)-H(13D)	109.2
C(3)-C(4)-C(2)	111.4(2)	C(14)-C(13)-H(13D)	109.2
O(2)-C(5)-O(1)	126.35(18)	C(12)-C(13)-H(13C)	109.2
O(2)-C(5)-N(1)	124.1(2)	C(14)-C(13)-H(13C)	109.2
O(1)-C(5)-N(1)	109.52(17)	H(13D)-C(13)-H(13C)	107.9
N(1)-C(6)-C(8)	110.60(16)	C(9)-C(14)-C(13)	109.05(15)
N(1)-C(6)-C(7)	109.25(16)	C(9)-C(14)-C(15)	110.80(15)
C(8)-C(6)-C(7)	109.86(16)	C(13)-C(14)-C(15)	113.87(16)
N(1)-C(6)-H(6)	109.0	C(9)-C(14)-H(14A)	107.6

C(13)-C(14)-H(14A)	107.6	C(24)-C(25)-C(26)	110.97(18)
C(15)-C(14)-H(14A)	107.6	C(24)-C(25)-H(25C)	109.4
C(18)-C(15)-C(16)	108.80(15)	C(26)-C(25)-H(25C)	109.4
C(18)-C(15)-C(14)	111.72(16)	C(24)-C(25)-H(25D)	109.4
C(16)-C(15)-C(14)	114.52(16)	C(26)-C(25)-H(25D)	109.4
C(18)-C(15)-H(15A)	107.1	H(25C)-C(25)-H(25D)	108.0
C(16)-C(15)-H(15A)	107.1	C(25)-C(26)-C(27)	111.47(18)
C(14)-C(15)-H(15A)	107.1	C(25)-C(26)-H(26D)	109.3
C(17)-C(16)-C(15)	112.08(18)	C(27)-C(26)-H(26D)	109.3
C(17)-C(16)-H(16C)	109.2	C(25)-C(26)-H(26C)	109.3
C(15)-C(16)-H(16C)	109.2	C(27)-C(26)-H(26C)	109.3
C(17)-C(16)-H(16D)	109.2	H(26D)-C(26)-H(26C)	108.0
C(15)-C(16)-H(16D)	109.2	C(26)-C(27)-C(22)	109.66(17)
H(16C)-C(16)-H(16D)	107.9	C(26)-C(27)-C(28)	112.42(17)
C(16)-C(17)-H(17D)	109.5	C(22)-C(27)-C(28)	112.60(16)
C(16)-C(17)-H(17F)	109.5	C(26)-C(27)-H(27A)	107.3
H(17D)-C(17)-H(17F)	109.5	C(22)-C(27)-H(27A)	107.3
C(16)-C(17)-H(17E)	109.5	C(28)-C(27)-H(27A)	107.3
H(17D)-C(17)-H(17E)	109.5	C(31)-C(28)-C(29)	110.83(17)
H(17F)-C(17)-H(17E)	109.5	C(31)-C(28)-C(27)	109.25(15)
O(4)-C(18)-N(3)	122.32(17)	C(29)-C(28)-C(27)	114.70(17)
O(4)-C(18)-C(15)	122.77(18)	C(31)-C(28)-H(28A)	107.2
N(3)-C(18)-C(15)	114.87(17)	C(29)-C(28)-H(28A)	107.2
N(3)-C(19)-C(20)	109.93(16)	C(27)-C(28)-H(28A)	107.2
N(3)-C(19)-C(21)	110.78(15)	C(30)-C(29)-C(28)	112.03(18)
C(20)-C(19)-C(21)	109.84(16)	C(30)-C(29)-H(29C)	109.2
N(3)-C(19)-H(19A)	108.7	C(28)-C(29)-H(29C)	109.2
C(20)-C(19)-H(19A)	108.7	C(30)-C(29)-H(29D)	109.2
C(21)-C(19)-H(19A)	108.7	C(28)-C(29)-H(29D)	109.2
C(19)-C(20)-H(20D)	109.5	H(29C)-C(29)-H(29D)	107.9
C(19)-C(20)-H(20F)	109.5	C(29)-C(30)-H(30D)	109.5
H(20D)-C(20)-H(20F)	109.5	C(29)-C(30)-H(30F)	109.5
C(19)-C(20)-H(20E)	109.5	H(30D)-C(30)-H(30F)	109.5
H(20D)-C(20)-H(20E)	109.5	C(29)-C(30)-H(30E)	109.5
H(20F)-C(20)-H(20E)	109.5	H(30D)-C(30)-H(30E)	109.5
O(5)-C(21)-N(4)	124.77(18)	H(30F)-C(30)-H(30E)	109.5
O(5)-C(21)-C(19)	121.00(17)	O(6)-C(31)-O(7)	122.5(2)
N(4)-C(21)-C(19)	114.17(17)	O(6)-C(31)-C(28)	126.6(2)
N(4)-C(22)-C(23)	109.81(17)	O(7)-C(31)-C(28)	110.88(17)
N(4)-C(22)-C(27)	110.51(16)	O(7)-C(32)-C(33)	109.72(18)
C(23)-C(22)-C(27)	110.18(16)	O(7)-C(32)-H(32F)	109.7
N(4)-C(22)-H(22A)	108.8	C(33)-C(32)-H(32F)	109.7
C(23)-C(22)-H(22A)	108.8	O(7)-C(32)-H(32E)	109.7
C(27)-C(22)-H(22A)	108.8	C(33)-C(32)-H(32E)	109.7
C(24)-C(23)-C(22)	111.74(17)	H(32F)-C(32)-H(32E)	108.2
C(24)-C(23)-H(23C)	109.3	C(38)-C(33)-C(34)	118.3(2)
C(22)-C(23)-H(23C)	109.3	C(38)-C(33)-C(32)	120.9(2)
C(24)-C(23)-H(23D)	109.3	C(34)-C(33)-C(32)	120.8(2)
C(22)-C(23)-H(23D)	109.3	C(35)-C(34)-C(33)	120.3(3)
H(23C)-C(23)-H(23D)	107.9	C(35)-C(34)-H(34B)	119.8
C(25)-C(24)-C(23)	111.31(18)	C(33)-C(34)-H(34B)	119.8
C(25)-C(24)-H(24C)	109.4	C(34)-C(35)-C(36)	120.1(3)
C(23)-C(24)-H(24C)	109.4	C(34)-C(35)-H(35B)	120.0
C(25)-C(24)-H(24D)	109.4	C(36)-C(35)-H(35B)	120.0
C(23)-C(24)-H(24D)	109.4	C(37)-C(36)-C(35)	119.7(3)
H(24C)-C(24)-H(24D)	108.0	C(37)-C(36)-H(36B)	120.2

C(35)-C(36)-H(36B)	120.2	H(7AC)-C(7A)-H(7AB)	109.5
C(36)-C(37)-C(38)	120.2(3)	C(6A)-C(7A)-H(7AA)	109.5
C(36)-C(37)-H(37B)	119.9	H(7AC)-C(7A)-H(7AA)	109.5
C(38)-C(37)-H(37B)	119.9	H(7AB)-C(7A)-H(7AA)	109.5
C(37)-C(38)-C(33)	121.3(3)	O(3A)-C(8A)-N(2A)	123.08(18)
C(37)-C(38)-H(38B)	119.3	O(3A)-C(8A)-C(6A)	121.61(18)
C(33)-C(38)-H(38B)	119.3	N(2A)-C(8A)-C(6A)	115.29(17)
C(5A)-O(1A)-C(4A)	120.96(16)	N(2A)-C(9A)-C(10A)	108.90(16)
C(31A)-O(7A)-C(32A)	117.87(16)	N(2A)-C(9A)-C(14A)	111.10(15)
C(5A)-N(1A)-C(6A)	120.37(17)	C(10A)-C(9A)-C(14A)	111.91(16)
C(5A)-N(1A)-H(1A)	115.1(17)	N(2A)-C(9A)-H(9A)	108.3
C(6A)-N(1A)-H(1A)	120.5(16)	C(10A)-C(9A)-H(9A)	108.3
C(8A)-N(2A)-C(9A)	123.69(17)	C(14A)-C(9A)-H(9A)	108.3
C(8A)-N(2A)-H(2A)	119.8(15)	C(11A)-C(10A)-C(9A)	112.02(18)
C(9A)-N(2A)-H(2A)	115.8(15)	C(11A)-C(10A)-H(10B)	109.2
C(18A)-N(3A)-C(19A)	120.52(17)	C(9A)-C(10A)-H(10B)	109.2
C(18A)-N(3A)-H(3A)	121.8(15)	C(11A)-C(10A)-H(10A)	109.2
C(19A)-N(3A)-H(3A)	117.7(15)	C(9A)-C(10A)-H(10A)	109.2
C(21A)-N(4A)-C(22A)	124.22(17)	H(10B)-C(10A)-H(10A)	107.9
C(21A)-N(4A)-H(4A)	116.4(14)	C(10A)-C(11A)-C(12A)	109.85(18)
C(22A)-N(4A)-H(4A)	119.4(14)	C(10A)-C(11A)-H(11A)	109.7
C(4A)-C(1A)-H(1AB)	109.5	C(12A)-C(11A)-H(11A)	109.7
C(4A)-C(1A)-H(1AC)	109.5	C(10A)-C(11A)-H(11B)	109.7
H(1AB)-C(1A)-H(1AC)	109.5	C(12A)-C(11A)-H(11B)	109.7
C(4A)-C(1A)-H(1AA)	109.5	H(11A)-C(11A)-H(11B)	108.2
H(1AB)-C(1A)-H(1AA)	109.5	C(13A)-C(12A)-C(11A)	110.19(18)
H(1AC)-C(1A)-H(1AA)	109.5	C(13A)-C(12A)-H(12A)	109.6
C(4A)-C(2A)-H(2AA)	109.5	C(11A)-C(12A)-H(12A)	109.6
C(4A)-C(2A)-H(2AC)	109.5	C(13A)-C(12A)-H(12B)	109.6
H(2AA)-C(2A)-H(2AC)	109.5	C(11A)-C(12A)-H(12B)	109.6
C(4A)-C(2A)-H(2AB)	109.5	H(12A)-C(12A)-H(12B)	108.1
H(2AA)-C(2A)-H(2AB)	109.5	C(12A)-C(13A)-C(14A)	112.08(18)
H(2AC)-C(2A)-H(2AB)	109.5	C(12A)-C(13A)-H(13A)	109.2
C(4A)-C(3A)-H(3AA)	109.5	C(14A)-C(13A)-H(13A)	109.2
C(4A)-C(3A)-H(3AC)	109.5	C(12A)-C(13A)-H(13B)	109.2
H(3AA)-C(3A)-H(3AC)	109.5	C(14A)-C(13A)-H(13B)	109.2
C(4A)-C(3A)-H(3AB)	109.5	H(13A)-C(13A)-H(13B)	107.9
H(3AA)-C(3A)-H(3AB)	109.5	C(13A)-C(14A)-C(9A)	109.63(16)
H(3AC)-C(3A)-H(3AB)	109.5	C(13A)-C(14A)-C(15A)	113.84(16)
O(1A)-C(4A)-C(3A)	110.09(19)	C(9A)-C(14A)-C(15A)	110.54(15)
O(1A)-C(4A)-C(2A)	102.29(17)	C(13A)-C(14A)-H(14)	107.5
C(3A)-C(4A)-C(2A)	111.0(2)	C(9A)-C(14A)-H(14)	107.5
O(1A)-C(4A)-C(1A)	109.66(18)	C(15A)-C(14A)-H(14)	107.5
C(3A)-C(4A)-C(1A)	113.00(19)	C(18A)-C(15A)-C(16A)	108.19(15)
C(2A)-C(4A)-C(1A)	110.3(2)	C(18A)-C(15A)-C(14A)	111.75(16)
O(2A)-C(5A)-O(1A)	126.32(18)	C(16A)-C(15A)-C(14A)	114.77(16)
O(2A)-C(5A)-N(1A)	123.9(2)	C(18A)-C(15A)-H(15)	107.3
O(1A)-C(5A)-N(1A)	109.79(17)	C(16A)-C(15A)-H(15)	107.3
N(1A)-C(6A)-C(8A)	110.75(17)	C(14A)-C(15A)-H(15)	107.3
N(1A)-C(6A)-C(7A)	109.50(16)	C(17A)-C(16A)-C(15A)	112.21(18)
C(8A)-C(6A)-C(7A)	109.65(16)	C(17A)-C(16A)-H(16B)	109.2
N(1A)-C(6A)-H(6A)	109.0	C(15A)-C(16A)-H(16B)	109.2
C(8A)-C(6A)-H(6A)	109.0	C(17A)-C(16A)-H(16A)	109.2
C(7A)-C(6A)-H(6A)	109.0	C(15A)-C(16A)-H(16A)	109.2
C(6A)-C(7A)-H(7AC)	109.5	H(16B)-C(16A)-H(16A)	107.9
C(6A)-C(7A)-H(7AB)	109.5	C(16A)-C(17A)-H(17B)	109.5

C(16A)-C(17A)-H(17A)	109.5	C(26A)-C(27A)-H(27)	107.6
H(17B)-C(17A)-H(17A)	109.5	C(22A)-C(27A)-H(27)	107.6
C(16A)-C(17A)-H(17C)	109.5	C(28A)-C(27A)-H(27)	107.6
H(17B)-C(17A)-H(17C)	109.5	C(31A)-C(28A)-C(29A)	110.31(17)
H(17A)-C(17A)-H(17C)	109.5	C(31A)-C(28A)-C(27A)	109.41(16)
O(4A)-C(18A)-N(3A)	121.73(17)	C(29A)-C(28A)-C(27A)	114.83(17)
O(4A)-C(18A)-C(15A)	122.78(18)	C(31A)-C(28A)-H(28)	107.3
N(3A)-C(18A)-C(15A)	115.42(17)	C(29A)-C(28A)-H(28)	107.3
N(3A)-C(19A)-C(20A)	110.24(16)	C(27A)-C(28A)-H(28)	107.3
N(3A)-C(19A)-C(21A)	110.85(15)	C(30A)-C(29A)-C(28A)	111.41(18)
C(20A)-C(19A)-C(21A)	110.02(16)	C(30A)-C(29A)-H(29A)	109.3
N(3A)-C(19A)-H(19)	108.6	C(28A)-C(29A)-H(29A)	109.3
C(20A)-C(19A)-H(19)	108.6	C(30A)-C(29A)-H(29B)	109.3
C(21A)-C(19A)-H(19)	108.6	C(28A)-C(29A)-H(29B)	109.3
C(19A)-C(20A)-H(20A)	109.5	H(29A)-C(29A)-H(29B)	108.0
C(19A)-C(20A)-H(20C)	109.5	C(29A)-C(30A)-H(30A)	109.5
H(20A)-C(20A)-H(20C)	109.5	C(29A)-C(30A)-H(30C)	109.5
C(19A)-C(20A)-H(20B)	109.5	H(30A)-C(30A)-H(30C)	109.5
H(20A)-C(20A)-H(20B)	109.5	C(29A)-C(30A)-H(30B)	109.5
H(20C)-C(20A)-H(20B)	109.5	H(30A)-C(30A)-H(30B)	109.5
O(5A)-C(21A)-N(4A)	124.52(18)	H(30C)-C(30A)-H(30B)	109.5
O(5A)-C(21A)-C(19A)	120.93(17)	O(6A)-C(31A)-O(7A)	122.7(2)
N(4A)-C(21A)-C(19A)	114.51(16)	O(6A)-C(31A)-C(28A)	126.54(19)
N(4A)-C(22A)-C(23A)	109.79(16)	O(7A)-C(31A)-C(28A)	110.76(17)
N(4A)-C(22A)-C(27A)	110.60(16)	O(7A)-C(32A)-C(33A)	112.17(19)
C(23A)-C(22A)-C(27A)	110.95(16)	O(7A)-C(32A)-C(33B)	104.14(19)
N(4A)-C(22A)-H(22)	108.5	C(33A)-C(32A)-C(33B)	8.03(19)
C(23A)-C(22A)-H(22)	108.5	O(7A)-C(32A)-H(32A)	109.2
C(27A)-C(22A)-H(22)	108.5	C(33A)-C(32A)-H(32A)	109.2
C(24A)-C(23A)-C(22A)	111.22(18)	C(33B)-C(32A)-H(32A)	113.1
C(24A)-C(23A)-H(23A)	109.4	O(7A)-C(32A)-H(32B)	109.2
C(22A)-C(23A)-H(23A)	109.4	C(33A)-C(32A)-H(32B)	109.2
C(24A)-C(23A)-H(23B)	109.4	C(33B)-C(32A)-H(32B)	113.2
C(22A)-C(23A)-H(23B)	109.4	H(32A)-C(32A)-H(32B)	107.9
H(23A)-C(23A)-H(23B)	108.0	O(7A)-C(32A)-H(32C)	110.9
C(25A)-C(24A)-C(23A)	110.54(18)	C(33A)-C(32A)-H(32C)	106.9
C(25A)-C(24A)-H(24B)	109.5	C(33B)-C(32A)-H(32C)	110.9
C(23A)-C(24A)-H(24B)	109.5	H(32A)-C(32A)-H(32C)	2.4
C(25A)-C(24A)-H(24A)	109.5	H(32B)-C(32A)-H(32C)	108.4
C(23A)-C(24A)-H(24A)	109.5	O(7A)-C(32A)-H(32D)	110.9
H(24B)-C(24A)-H(24A)	108.1	C(33A)-C(32A)-H(32D)	106.8
C(26A)-C(25A)-C(24A)	110.82(17)	C(33B)-C(32A)-H(32D)	110.9
C(26A)-C(25A)-H(25B)	109.5	H(32A)-C(32A)-H(32D)	108.5
C(24A)-C(25A)-H(25B)	109.5	H(32B)-C(32A)-H(32D)	2.4
C(26A)-C(25A)-H(25A)	109.5	H(32C)-C(32A)-H(32D)	108.9
C(24A)-C(25A)-H(25A)	109.5	C(34A)-C(33A)-C(38A)	120.0
H(25B)-C(25A)-H(25A)	108.1	C(34A)-C(33A)-C(32A)	117.1(2)
C(25A)-C(26A)-C(27A)	112.18(18)	C(38A)-C(33A)-C(32A)	122.9(2)
C(25A)-C(26A)-H(26B)	109.2	C(35A)-C(34A)-C(33A)	120.0
C(27A)-C(26A)-H(26B)	109.2	C(35A)-C(34A)-H(34)	120.0
C(25A)-C(26A)-H(26A)	109.2	C(33A)-C(34A)-H(34)	120.0
C(27A)-C(26A)-H(26A)	109.2	C(36A)-C(35A)-C(34A)	120.0
H(26B)-C(26A)-H(26A)	107.9	C(36A)-C(35A)-H(35)	120.0
C(26A)-C(27A)-C(22A)	110.16(17)	C(34A)-C(35A)-H(35)	120.0
C(26A)-C(27A)-C(28A)	112.42(17)	C(37A)-C(36A)-C(35A)	120.0
C(22A)-C(27A)-C(28A)	111.36(15)	C(37A)-C(36A)-H(36)	120.0

C(35A)-C(36A)-H(36)	120.0	C(36B)-C(35B)-C(34B)	120.0
C(36A)-C(37A)-C(38A)	120.0	C(36B)-C(35B)-H(35A)	120.0
C(36A)-C(37A)-H(37)	120.0	C(34B)-C(35B)-H(35A)	120.0
C(38A)-C(37A)-H(37)	120.0	C(37B)-C(36B)-C(35B)	120.0
C(37A)-C(38A)-C(33A)	120.0	C(37B)-C(36B)-H(36A)	120.0
C(37A)-C(38A)-H(38)	120.0	C(35B)-C(36B)-H(36A)	120.0
C(33A)-C(38A)-H(38)	120.0	C(36B)-C(37B)-C(38B)	120.0
C(34B)-C(33B)-C(38B)	120.0	C(36B)-C(37B)-H(37A)	120.0
C(34B)-C(33B)-C(32A)	123.89(19)	C(38B)-C(37B)-H(37A)	120.0
C(38B)-C(33B)-C(32A)	116.11(19)	C(37B)-C(38B)-C(33B)	120.0
C(33B)-C(34B)-C(35B)	120.0	C(37B)-C(38B)-H(38A)	120.0
C(33B)-C(34B)-H(34A)	120.0	C(33B)-C(38B)-H(38A)	120.0
C(35B)-C(34B)-H(34A)	120.0	H(1W)-O(1W)-H(2W)	104.5

Symmetry transformations used to generate equivalent atoms:

Table S38. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for gellman124a. The anisotropic displacement factor exponent takes the form: $-2\pi^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
O(1)	56(1)	26(1)	32(1)	-3(1)	4(1)	0(1)
O(2)	46(1)	24(1)	38(1)	2(1)	2(1)	-3(1)
O(3)	25(1)	27(1)	40(1)	1(1)	4(1)	-2(1)
O(4)	36(1)	23(1)	38(1)	3(1)	-2(1)	-1(1)
O(5)	30(1)	28(1)	43(1)	1(1)	3(1)	4(1)
O(6)	58(1)	41(1)	35(1)	-8(1)	3(1)	2(1)
O(7)	38(1)	38(1)	32(1)	-2(1)	-2(1)	1(1)
N(1)	43(1)	23(1)	30(1)	2(1)	1(1)	-4(1)
N(2)	24(1)	26(1)	33(1)	-2(1)	-1(1)	2(1)
N(3)	30(1)	21(1)	32(1)	1(1)	0(1)	0(1)
N(4)	24(1)	23(1)	38(1)	1(1)	3(1)	1(1)
C(1)	48(2)	40(1)	52(2)	3(1)	3(1)	7(1)
C(2)	62(2)	43(1)	38(1)	-8(1)	2(1)	4(1)
C(3)	53(2)	43(1)	54(2)	-13(1)	-4(1)	-2(1)
C(4)	45(1)	29(1)	39(1)	-7(1)	-2(1)	3(1)
C(5)	38(1)	28(1)	31(1)	-2(1)	-4(1)	-3(1)
C(6)	32(1)	24(1)	31(1)	1(1)	-1(1)	0(1)
C(7)	42(1)	27(1)	40(1)	0(1)	-3(1)	4(1)
C(8)	30(1)	16(1)	33(1)	-2(1)	0(1)	-1(1)
C(9)	28(1)	23(1)	31(1)	-2(1)	-2(1)	1(1)
C(10)	43(1)	27(1)	36(1)	-4(1)	-2(1)	6(1)
C(11)	53(1)	33(1)	33(1)	-5(1)	-3(1)	2(1)
C(12)	49(1)	33(1)	31(1)	1(1)	-8(1)	0(1)
C(13)	40(1)	27(1)	36(1)	2(1)	-3(1)	2(1)
C(14)	26(1)	23(1)	30(1)	0(1)	0(1)	-1(1)
C(15)	29(1)	21(1)	34(1)	1(1)	0(1)	-2(1)
C(16)	33(1)	26(1)	47(1)	-3(1)	5(1)	-5(1)
C(17)	34(1)	37(1)	58(2)	-3(1)	14(1)	0(1)
C(18)	25(1)	24(1)	32(1)	-1(1)	6(1)	0(1)
C(19)	27(1)	23(1)	37(1)	-2(1)	3(1)	0(1)
C(20)	42(1)	32(1)	39(1)	-5(1)	4(1)	-7(1)
C(21)	24(1)	21(1)	33(1)	-4(1)	-3(1)	-2(1)
C(22)	27(1)	25(1)	40(1)	0(1)	2(1)	-1(1)
C(23)	34(1)	27(1)	44(1)	-2(1)	-4(1)	-6(1)
C(24)	39(1)	24(1)	54(1)	3(1)	-2(1)	-10(1)
C(25)	50(1)	27(1)	43(1)	2(1)	4(1)	-10(1)
C(26)	47(1)	26(1)	42(1)	0(1)	5(1)	-6(1)
C(27)	32(1)	22(1)	38(1)	-1(1)	3(1)	-2(1)
C(28)	35(1)	22(1)	36(1)	0(1)	3(1)	-3(1)
C(29)	42(1)	28(1)	53(1)	-7(1)	2(1)	0(1)
C(30)	46(1)	26(1)	60(2)	-6(1)	2(1)	2(1)
C(31)	38(1)	21(1)	35(1)	-1(1)	5(1)	-8(1)
C(32)	46(1)	44(1)	34(1)	1(1)	-8(1)	0(1)
C(33)	44(1)	40(1)	39(1)	-12(1)	-3(1)	0(1)
C(34)	46(2)	37(1)	102(2)	-14(1)	12(2)	-3(1)
C(35)	57(2)	46(2)	126(3)	-13(2)	13(2)	-16(1)
C(36)	47(2)	78(2)	77(2)	-22(2)	5(1)	-19(2)
C(37)	37(2)	89(2)	59(2)	-6(2)	-7(1)	5(1)
C(38)	48(2)	58(2)	51(2)	0(1)	-10(1)	8(1)
O(1A)	63(1)	24(1)	29(1)	2(1)	-1(1)	0(1)
O(2A)	47(1)	23(1)	36(1)	-3(1)	-1(1)	-1(1)

O(3A)	30(1)	26(1)	39(1)	-1(1)	-4(1)	-2(1)
O(4A)	36(1)	25(1)	40(1)	-5(1)	4(1)	-1(1)
O(5A)	26(1)	29(1)	47(1)	-5(1)	-3(1)	3(1)
O(6A)	54(1)	49(1)	31(1)	1(1)	-2(1)	12(1)
O(7A)	34(1)	37(1)	33(1)	3(1)	3(1)	2(1)
N(1A)	44(1)	23(1)	30(1)	-1(1)	-1(1)	-2(1)
N(2A)	25(1)	25(1)	34(1)	1(1)	0(1)	1(1)
N(3A)	29(1)	21(1)	34(1)	-3(1)	1(1)	0(1)
N(4A)	25(1)	22(1)	36(1)	0(1)	-2(1)	2(1)
C(1A)	49(2)	37(1)	52(2)	0(1)	-2(1)	4(1)
C(2A)	84(2)	39(1)	44(1)	8(1)	-7(1)	5(1)
C(3A)	57(2)	40(1)	50(2)	12(1)	9(1)	-5(1)
C(4A)	52(1)	25(1)	37(1)	6(1)	0(1)	2(1)
C(5A)	35(1)	25(1)	34(1)	-1(1)	6(1)	-3(1)
C(6A)	31(1)	25(1)	33(1)	-1(1)	2(1)	-1(1)
C(7A)	43(1)	28(1)	41(1)	0(1)	4(1)	6(1)
C(8A)	30(1)	14(1)	36(1)	3(1)	0(1)	-1(1)
C(9A)	29(1)	25(1)	33(1)	1(1)	1(1)	0(1)
C(10A)	36(1)	34(1)	38(1)	7(1)	-3(1)	0(1)
C(11A)	47(1)	36(1)	33(1)	8(1)	-1(1)	-3(1)
C(12A)	42(1)	44(1)	31(1)	0(1)	2(1)	-7(1)
C(13A)	37(1)	34(1)	34(1)	-5(1)	4(1)	-4(1)
C(14A)	27(1)	23(1)	33(1)	-1(1)	-1(1)	-3(1)
C(15A)	28(1)	24(1)	31(1)	-2(1)	0(1)	0(1)
C(16A)	28(1)	24(1)	49(1)	0(1)	-4(1)	-3(1)
C(17A)	33(1)	32(1)	56(1)	2(1)	-11(1)	-2(1)
C(18A)	23(1)	26(1)	33(1)	-3(1)	-4(1)	3(1)
C(19A)	28(1)	22(1)	36(1)	0(1)	0(1)	1(1)
C(20A)	37(1)	29(1)	39(1)	2(1)	-2(1)	-5(1)
C(21A)	25(1)	19(1)	35(1)	2(1)	1(1)	-1(1)
C(22A)	28(1)	23(1)	40(1)	-2(1)	-2(1)	-1(1)
C(23A)	35(1)	25(1)	45(1)	0(1)	6(1)	-4(1)
C(24A)	34(1)	24(1)	58(2)	-5(1)	1(1)	-8(1)
C(25A)	43(1)	28(1)	48(1)	-5(1)	-6(1)	-4(1)
C(26A)	43(1)	31(1)	39(1)	-4(1)	-4(1)	-2(1)
C(27A)	32(1)	24(1)	37(1)	-1(1)	-2(1)	0(1)
C(28A)	33(1)	24(1)	34(1)	0(1)	0(1)	0(1)
C(29A)	36(1)	28(1)	47(1)	3(1)	1(1)	3(1)
C(30A)	47(1)	28(1)	47(1)	4(1)	4(1)	2(1)
C(31A)	38(1)	21(1)	36(1)	-3(1)	-3(1)	1(1)
C(32A)	40(1)	41(1)	36(1)	2(1)	7(1)	6(1)
C(33A)	32(1)	43(1)	37(2)	6(1)	8(1)	8(1)
C(34A)	38(1)	48(1)	43(1)	4(1)	2(1)	7(1)
C(35A)	36(1)	70(1)	63(2)	2(1)	-2(1)	1(1)
C(36A)	44(2)	76(2)	77(4)	-5(3)	-4(2)	-16(2)
C(37A)	36(1)	70(1)	63(2)	2(1)	-2(1)	1(1)
C(38A)	38(1)	48(1)	43(1)	4(1)	2(1)	7(1)
C(33B)	32(1)	43(1)	37(2)	6(1)	8(1)	8(1)
C(34B)	38(1)	48(1)	43(1)	4(1)	2(1)	7(1)
C(35B)	36(1)	70(1)	63(2)	2(1)	-2(1)	1(1)
C(36B)	44(2)	76(2)	77(4)	-5(3)	-4(2)	-16(2)
C(37B)	36(1)	70(1)	63(2)	2(1)	-2(1)	1(1)
C(38B)	38(1)	48(1)	43(1)	4(1)	2(1)	7(1)
O(1W)	75(1)	39(1)	55(1)	0(1)	4(1)	13(1)

Table S39. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for gellman124a.

	x	y	z	U(eq)
H(1)	-1024(15)	-8116(14)	-1222(8)	39
H(2)	-1713(15)	-8355(12)	264(8)	33
H(3)	-1152(14)	-10494(13)	-215(8)	33
H(4)	-1226(15)	-12650(13)	516(8)	34
H(1B)	436	-10205	-1356	70
H(1D)	40	-10995	-1534	70
H(1C)	-340	-10548	-1090	70
H(2C)	-537	-9561	-2397	71
H(2D)	-34	-10346	-2347	71
H(2B)	309	-9565	-2121	71
H(3C)	-1693	-10539	-1518	75
H(3D)	-1307	-10948	-1974	75
H(3B)	-1754	-10134	-2027	75
H(6)	-2083	-8412	-511	35
H(7B)	-2017	-7082	-340	54
H(7C)	-2193	-7183	-895	54
H(7A)	-1301	-6968	-716	54
H(9)	-128	-8219	541	33
H(10C)	-1033	-7382	927	42
H(10D)	-1588	-8090	1090	42
H(11D)	-801	-7703	1738	47
H(11C)	13	-7784	1433	47
H(12D)	-1010	-9047	1709	45
H(12C)	-122	-8887	1916	45
H(13D)	-70	-9871	1330	41
H(13C)	457	-9144	1166	41
H(14A)	-1155	-9468	840	32
H(15A)	-459	-9524	54	33
H(16C)	875	-9230	328	42
H(16D)	892	-10033	608	42
H(17D)	746	-9916	-402	65
H(17F)	799	-10712	-117	65
H(17E)	1564	-10147	-133	65
H(19A)	-778	-11934	-18	35
H(20D)	-2036	-11361	-623	56
H(20F)	-1782	-12250	-598	56
H(20E)	-1156	-11624	-796	56
H(22A)	-2716	-12426	970	37
H(23C)	-2835	-13519	474	42
H(23D)	-2044	-13895	706	42
H(24C)	-3551	-13649	1187	47
H(24D)	-3187	-14474	1047	47
H(25C)	-2135	-14333	1596	48
H(25D)	-2975	-14195	1867	48
H(26D)	-2016	-13231	2077	46
H(26C)	-2793	-12851	1831	46
H(27A)	-1304	-13173	1348	36
H(28A)	-1098	-11821	1291	37
H(29C)	-2482	-11411	1430	49
H(29D)	-2336	-11566	1984	49

H(30D)	-1421	-10473	1440	65
H(30F)	-1307	-10615	1997	65
H(30E)	-2127	-10241	1801	65
H(32F)	511	-13297	2213	50
H(32E)	217	-12558	2508	50
H(34B)	655	-11305	1883	74
H(35B)	1848	-10681	1679	92
H(36B)	3087	-11313	1762	81
H(37B)	3115	-12585	2000	74
H(38B)	1930	-13213	2197	63
H(1A)	-4395(15)	-11802(14)	-1239(8)	39
H(2A)	-3837(15)	-11610(13)	279(8)	33
H(3A)	-4289(14)	-9434(13)	-198(8)	34
H(4A)	-4248(15)	-7291(13)	558(8)	34
H(1AB)	-5061	-9333	-1092	69
H(1AC)	-5766	-9672	-1421	69
H(1AA)	-5342	-8873	-1556	69
H(2AA)	-5011	-9433	-2365	84
H(2AC)	-5454	-10210	-2203	84
H(2AB)	-4541	-10238	-2392	84
H(3AA)	-3855	-8921	-1862	74
H(3AC)	-3425	-9747	-1889	74
H(3AB)	-3622	-9368	-1384	74
H(6A)	-3414	-11564	-482	36
H(7AC)	-4198	-12980	-742	56
H(7AB)	-3511	-12903	-345	56
H(7AA)	-3292	-12763	-892	56
H(9A)	-5421	-11726	529	35
H(10B)	-4598	-12607	936	43
H(10A)	-4017	-11928	1115	43
H(11A)	-4893	-12275	1738	46
H(11B)	-5668	-12132	1407	46
H(12A)	-5475	-11042	1903	47
H(12B)	-4568	-10943	1711	47
H(13A)	-5432	-10062	1316	42
H(13B)	-5997	-10757	1142	42
H(14)	-4353	-10524	847	33
H(15)	-4970	-10426	48	33
H(16B)	-6340	-10719	269	40
H(16A)	-6385	-9916	547	40
H(17B)	-6124	-10037	-454	61
H(17A)	-6201	-9239	-172	61
H(17C)	-6968	-9798	-218	61
H(19)	-4713	-8002	29	34
H(20A)	-3707	-7624	-538	53
H(20C)	-4334	-8234	-754	53
H(20B)	-3452	-8510	-593	53
H(22)	-2806	-7565	1044	36
H(23A)	-2609	-6462	566	42
H(23B)	-3392	-6058	790	42
H(24B)	-1936	-6407	1304	47
H(24A)	-2225	-5557	1160	47
H(25B)	-2544	-5822	1968	47
H(25A)	-3349	-5650	1670	47
H(26B)	-3564	-6745	2144	45
H(26A)	-2779	-7154	1927	45

H(27)	-4224	-6806	1405	37
H(28)	-4404	-8162	1323	37
H(29A)	-3003	-8528	1510	44
H(29B)	-3252	-8449	2057	44
H(30A)	-3940	-9517	1396	61
H(30C)	-4306	-9391	1916	61
H(30B)	-3415	-9738	1853	61
H(32A)	-6221	-6861	2178	47
H(32B)	-5846	-7522	2508	47
H(32C)	-6245	-6868	2179	47
H(32D)	-5868	-7535	2510	47
H(34)	-6100	-8903	2167	52
H(35)	-7170	-9755	2022	68
H(36)	-8456	-9275	1840	79
H(37)	-8672	-7943	1803	68
H(38)	-7602	-7091	1948	52
H(34A)	-5989	-8901	1866	52
H(35A)	-7049	-9695	1613	68
H(36A)	-8377	-9227	1609	79
H(37A)	-8645	-7966	1857	68
H(38A)	-7585	-7172	2111	52
H(1W)	-2830	-9566	541	84
H(2W)	-2654	-10423	553	84

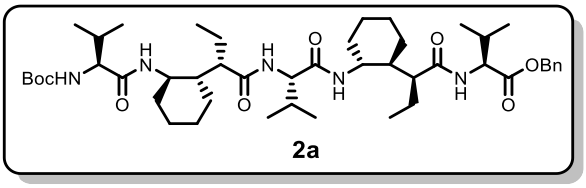
Table S40. Torsion angles [°] for gellman124a.

C(5)-O(1)-C(4)-C(1)	-62.0(3)	C(24)-C(25)-C(26)-C(27)	-56.6(3)
C(5)-O(1)-C(4)-C(3)	62.5(3)	C(25)-C(26)-C(27)-C(22)	57.7(2)
C(5)-O(1)-C(4)-C(2)	-179.26(19)	C(25)-C(26)-C(27)-C(28)	-176.19(18)
C(4)-O(1)-C(5)-O(2)	6.1(3)	N(4)-C(22)-C(27)-C(26)	-178.65(16)
C(4)-O(1)-C(5)-N(1)	-172.75(18)	C(23)-C(22)-C(27)-C(26)	-57.1(2)
C(6)-N(1)-C(5)-O(2)	12.2(3)	N(4)-C(22)-C(27)-C(28)	55.4(2)
C(6)-N(1)-C(5)-O(1)	-168.94(18)	C(23)-C(22)-C(27)-C(28)	176.93(17)
C(5)-N(1)-C(6)-C(8)	64.3(2)	C(26)-C(27)-C(28)-C(31)	66.2(2)
C(5)-N(1)-C(6)-C(7)	-174.62(19)	C(22)-C(27)-C(28)-C(31)	-169.32(16)
C(9)-N(2)-C(8)-O(3)	1.6(3)	C(26)-C(27)-C(28)-C(29)	-58.9(2)
C(9)-N(2)-C(8)-C(6)	-177.30(16)	C(22)-C(27)-C(28)-C(29)	65.6(2)
N(1)-C(6)-C(8)-O(3)	28.6(2)	C(31)-C(28)-C(29)-C(30)	68.0(2)
C(7)-C(6)-C(8)-O(3)	-92.0(2)	C(27)-C(28)-C(29)-C(30)	-167.67(19)
N(1)-C(6)-C(8)-N(2)	-152.47(16)	C(32)-O(7)-C(31)-O(6)	-2.4(3)
C(7)-C(6)-C(8)-N(2)	86.9(2)	C(32)-O(7)-C(31)-C(28)	178.49(16)
C(8)-N(2)-C(9)-C(10)	137.80(18)	C(29)-C(28)-C(31)-O(6)	12.1(3)
C(8)-N(2)-C(9)-C(14)	-99.3(2)	C(27)-C(28)-C(31)-O(6)	-115.2(2)
N(2)-C(9)-C(10)-C(11)	178.03(17)	C(29)-C(28)-C(31)-O(7)	-168.84(16)
C(14)-C(9)-C(10)-C(11)	55.4(2)	C(27)-C(28)-C(31)-O(7)	63.8(2)
C(9)-C(10)-C(11)-C(12)	-55.4(3)	C(31)-O(7)-C(32)-C(33)	-112.5(2)
C(10)-C(11)-C(12)-C(13)	56.0(3)	O(7)-C(32)-C(33)-C(38)	-134.9(2)
C(11)-C(12)-C(13)-C(14)	-58.0(2)	O(7)-C(32)-C(33)-C(34)	44.1(3)
N(2)-C(9)-C(14)-C(13)	-175.02(16)	C(38)-C(33)-C(34)-C(35)	-0.7(4)
C(10)-C(9)-C(14)-C(13)	-54.3(2)	C(32)-C(33)-C(34)-C(35)	-179.7(3)
N(2)-C(9)-C(14)-C(15)	58.9(2)	C(33)-C(34)-C(35)-C(36)	-0.8(5)
C(10)-C(9)-C(14)-C(15)	179.58(16)	C(34)-C(35)-C(36)-C(37)	2.1(5)
C(12)-C(13)-C(14)-C(9)	56.4(2)	C(35)-C(36)-C(37)-C(38)	-1.8(5)
C(12)-C(13)-C(14)-C(15)	-179.30(16)	C(36)-C(37)-C(38)-C(33)	0.2(4)
C(9)-C(14)-C(15)-C(18)	-150.29(16)	C(34)-C(33)-C(38)-C(37)	1.0(4)
C(13)-C(14)-C(15)-C(18)	86.3(2)	C(32)-C(33)-C(38)-C(37)	-179.9(2)
C(9)-C(14)-C(15)-C(16)	85.5(2)	C(5A)-O(1A)-C(4A)-C(3A)	59.1(3)
C(13)-C(14)-C(15)-C(16)	-37.9(2)	C(5A)-O(1A)-C(4A)-C(2A)	177.1(2)
C(18)-C(15)-C(16)-C(17)	61.2(2)	C(5A)-O(1A)-C(4A)-C(1A)	-65.9(3)
C(14)-C(15)-C(16)-C(17)	-173.04(17)	C(4A)-O(1A)-C(5A)-O(2A)	6.1(3)
C(19)-N(3)-C(18)-O(4)	-5.8(3)	C(4A)-O(1A)-C(5A)-N(1A)	-171.81(18)
C(19)-N(3)-C(18)-C(15)	171.79(16)	C(6A)-N(1A)-C(5A)-O(2A)	7.3(3)
C(16)-C(15)-C(18)-O(4)	61.1(2)	C(6A)-N(1A)-C(5A)-O(1A)	-174.76(18)
C(14)-C(15)-C(18)-O(4)	-66.3(2)	C(5A)-N(1A)-C(6A)-C(8A)	67.3(2)
C(16)-C(15)-C(18)-N(3)	-116.49(19)	C(5A)-N(1A)-C(6A)-C(7A)	-171.71(19)
C(14)-C(15)-C(18)-N(3)	116.10(19)	C(9A)-N(2A)-C(8A)-O(3A)	-2.2(3)
C(18)-N(3)-C(19)-C(20)	-165.70(18)	C(9A)-N(2A)-C(8A)-C(6A)	179.21(16)
C(18)-N(3)-C(19)-C(21)	72.7(2)	N(1A)-C(6A)-C(8A)-O(3A)	31.1(2)
C(22)-N(4)-C(21)-O(5)	-2.1(3)	C(7A)-C(6A)-C(8A)-O(3A)	-89.8(2)
C(22)-N(4)-C(21)-C(19)	-179.41(17)	N(1A)-C(6A)-C(8A)-N(2A)	-150.28(16)
N(3)-C(19)-C(21)-O(5)	57.9(2)	C(7A)-C(6A)-C(8A)-N(2A)	88.8(2)
C(20)-C(19)-C(21)-O(5)	-63.7(2)	C(8A)-N(2A)-C(9A)-C(10A)	136.98(18)
N(3)-C(19)-C(21)-N(4)	-124.68(18)	C(8A)-N(2A)-C(9A)-C(14A)	-99.3(2)
C(20)-C(19)-C(21)-N(4)	113.68(19)	N(2A)-C(9A)-C(10A)-C(11A)	178.37(17)
C(21)-N(4)-C(22)-C(23)	113.0(2)	C(14A)-C(9A)-C(10A)-C(11A)	55.2(2)
C(21)-N(4)-C(22)-C(27)	-125.29(19)	C(9A)-C(10A)-C(11A)-C(12A)	-56.7(2)
N(4)-C(22)-C(23)-C(24)	178.48(17)	C(10A)-C(11A)-C(12A)-C(13A)	57.8(2)
C(27)-C(22)-C(23)-C(24)	56.5(2)	C(11A)-C(12A)-C(13A)-C(14A)	-58.5(2)
C(22)-C(23)-C(24)-C(25)	-55.3(2)	C(12A)-C(13A)-C(14A)-C(9A)	55.5(2)
C(23)-C(24)-C(25)-C(26)	54.7(3)	C(12A)-C(13A)-C(14A)-C(15A)	179.86(16)

N(2A)-C(9A)-C(14A)-C(13A)	-175.24(16)
C(10A)-C(9A)-C(14A)-C(13A)	-53.3(2)
N(2A)-C(9A)-C(14A)-C(15A)	58.5(2)
C(10A)-C(9A)-C(14A)-C(15A)	-179.57(17)
C(13A)-C(14A)-C(15A)-C(18A)	82.1(2)
C(9A)-C(14A)-C(15A)-C(18A)	-153.96(16)
C(13A)-C(14A)-C(15A)-C(16A)	-41.5(2)
C(9A)-C(14A)-C(15A)-C(16A)	82.4(2)
C(18A)-C(15A)-C(16A)-C(17A)	62.1(2)
C(14A)-C(15A)-C(16A)-C(17A)	-172.38(17)
C(19A)-N(3A)-C(18A)-O(4A)	-6.0(3)
C(19A)-N(3A)-C(18A)-C(15A)	171.07(16)
C(16A)-C(15A)-C(18A)-O(4A)	64.1(2)
C(14A)-C(15A)-C(18A)-O(4A)	-63.2(2)
C(16A)-C(15A)-C(18A)-N(3A)	-113.01(19)
C(14A)-C(15A)-C(18A)-N(3A)	119.70(18)
C(18A)-N(3A)-C(19A)-C(20A)	-164.34(17)
C(18A)-N(3A)-C(19A)-C(21A)	73.6(2)
C(22A)-N(4A)-C(21A)-O(5A)	-3.6(3)
C(22A)-N(4A)-C(21A)-C(19A)	178.63(17)
N(3A)-C(19A)-C(21A)-O(5A)	55.8(2)
C(20A)-C(19A)-C(21A)-O(5A)	-66.4(2)
N(3A)-C(19A)-C(21A)-N(4A)	-126.34(18)
C(20A)-C(19A)-C(21A)-N(4A)	111.47(19)
C(21A)-N(4A)-C(22A)-C(23A)	111.5(2)
C(21A)-N(4A)-C(22A)-C(27A)	-125.69(19)
N(4A)-C(22A)-C(23A)-C(24A)	179.13(17)
C(27A)-C(22A)-C(23A)-C(24A)	56.6(2)
C(22A)-C(23A)-C(24A)-C(25A)	-57.1(2)
C(23A)-C(24A)-C(25A)-C(26A)	56.6(2)
C(24A)-C(25A)-C(26A)-C(27A)	-56.3(3)
C(25A)-C(26A)-C(27A)-C(22A)	55.2(2)
C(25A)-C(26A)-C(27A)-C(28A)	-179.99(18)
N(4A)-C(22A)-C(27A)-C(26A)	-176.89(15)
C(23A)-C(22A)-C(27A)-C(26A)	-54.8(2)
N(4A)-C(22A)-C(27A)-C(28A)	57.7(2)
C(23A)-C(22A)-C(27A)-C(28A)	179.77(17)
C(26A)-C(27A)-C(28A)-C(31A)	66.9(2)
C(22A)-C(27A)-C(28A)-C(31A)	-168.94(17)
C(26A)-C(27A)-C(28A)-C(29A)	-57.8(2)
C(22A)-C(27A)-C(28A)-C(29A)	66.4(2)
C(31A)-C(28A)-C(29A)-C(30A)	72.9(2)
C(27A)-C(28A)-C(29A)-C(30A)	-162.91(18)
C(32A)-O(7A)-C(31A)-O(6A)	-3.3(3)
C(32A)-O(7A)-C(31A)-C(28A)	176.56(16)
C(29A)-C(28A)-C(31A)-O(6A)	17.1(3)
C(27A)-C(28A)-C(31A)-O(6A)	-110.2(2)
C(29A)-C(28A)-C(31A)-O(7A)	-162.73(16)
C(27A)-C(28A)-C(31A)-O(7A)	70.0(2)
C(31A)-O(7A)-C(32A)-C(33A)	-109.3(2)
C(31A)-O(7A)-C(32A)-C(33B)	-109.4(3)
O(7A)-C(32A)-C(33A)-C(34A)	62.9(3)
C(33B)-C(32A)-C(33A)-C(34A)	63(2)
O(7A)-C(32A)-C(33A)-C(38A)	-119.0(2)
C(33B)-C(32A)-C(33A)-C(38A)	-119(2)
C(38A)-C(33A)-C(34A)-C(35A)	0.0
C(32A)-C(33A)-C(34A)-C(35A)	178.2(3)
C(33A)-C(34A)-C(35A)-C(36A)	0.0
C(34A)-C(35A)-C(36A)-C(37A)	0.0
C(35A)-C(36A)-C(37A)-C(38A)	0.0
C(36A)-C(37A)-C(38A)-C(33A)	0.0
C(34A)-C(33A)-C(38A)-C(37A)	0.0
C(32A)-C(33A)-C(38A)-C(37A)	-178.1(3)
O(7A)-C(32A)-C(33B)-C(34B)	43.4(4)
C(33A)-C(32A)-C(33B)-C(34B)	-136(3)
O(7A)-C(32A)-C(33B)-C(38B)	-136.83(13)
C(33A)-C(32A)-C(33B)-C(38B)	44(2)
C(38B)-C(33B)-C(34B)-C(35B)	0.0
C(32A)-C(33B)-C(34B)-C(35B)	179.8(5)
C(33B)-C(34B)-C(35B)-C(36B)	0.0
C(34B)-C(35B)-C(36B)-C(37B)	0.0
C(35B)-C(36B)-C(37B)-C(38B)	0.0
C(36B)-C(37B)-C(38B)-C(33B)	0.0
C(34B)-C(33B)-C(38B)-C(37B)	0.0
C(32A)-C(33B)-C(38B)-C(37B)	-179.8(5)

Symmetry transformations used to generate equivalent atoms:

Crystallographic Experimental Section for Compound 2a



2a

Data Collection

A colorless crystal with approximate dimensions 0.684 × 0.524 × 0.42 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker SMART APEXII diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 41 frames collected at intervals of 0.6º in a 25º range about ω with the exposure time of 1 second per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9984 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.8 Å. A total of 84596 data were harvested by collecting 16 sets of frames with 0.6º scans in ω and φ with an exposure time 2/6/10 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were uniquely consistent for the space group $P2_12_12_1$ that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms were refined with anisotropic displacement coefficients. All non-amide hydrogen atoms were included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients. The amide hydrogens were located in the difference Fourier map and refined independently.

There is one solvent accessible void in the asymmetric unit. The void is occupied by a heptane molecule 70.26(18)% of the time and a chloroform molecule the rest of the time. The solvent molecules were refined with six constraints and restraints.

The absolute configuration has been unequivocally established as follows: C9(S), C14(S), C17(S), C22(R), C24(S), C29(S), C32(S), C37(R), C39(S).

The final least-squares refinement of 669 parameters against 10935 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0315 and 0.0809, respectively. The final difference Fourier map was featureless.

Summary

Crystal Data for $C_{52.21542}H_{88.53855}Cl_{0.89229}N_5O_8$ (*M* =946.03): orthorhombic, space group $P2_12_12_1$ (no. 19), *a* = 12.0179(19) Å, *b* = 17.559(4) Å, *c* = 26.124(4) Å, *V* = 5512.9(16) Å^3, *Z* = 4, *T* = 100.01 K, μ(Cu Kα) = 0.988 mm^{-1}, *Dcalc* = 1.140 g/mm^3, 84596 reflections measured ($6.064 \leq 2\Theta \leq 146.28$), 10935 unique ($R_{int}$ = 0.0217) which were used in all calculations. The final R_1 was 0.0315 ($I > 2\sigma(I)$) and wR_2 was 0.0809 (all data).

References

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[2] Sheldrick, G. M. (2008) SHELXL. *Acta Cryst.* **A64**, 112-122.

[3] Dolomanov, O.V.; Bourhis, L.J.; Gildea, R.J.; Howard, J.A.K.; Puschmann, H. "OLEX2: a complete structure solution, refinement and analysis program". *J. Appl. Cryst.* (2009) **42**, *339-341*.

[4] Guzei, I.A. (2013). Internal laboratory computer programs Gn.

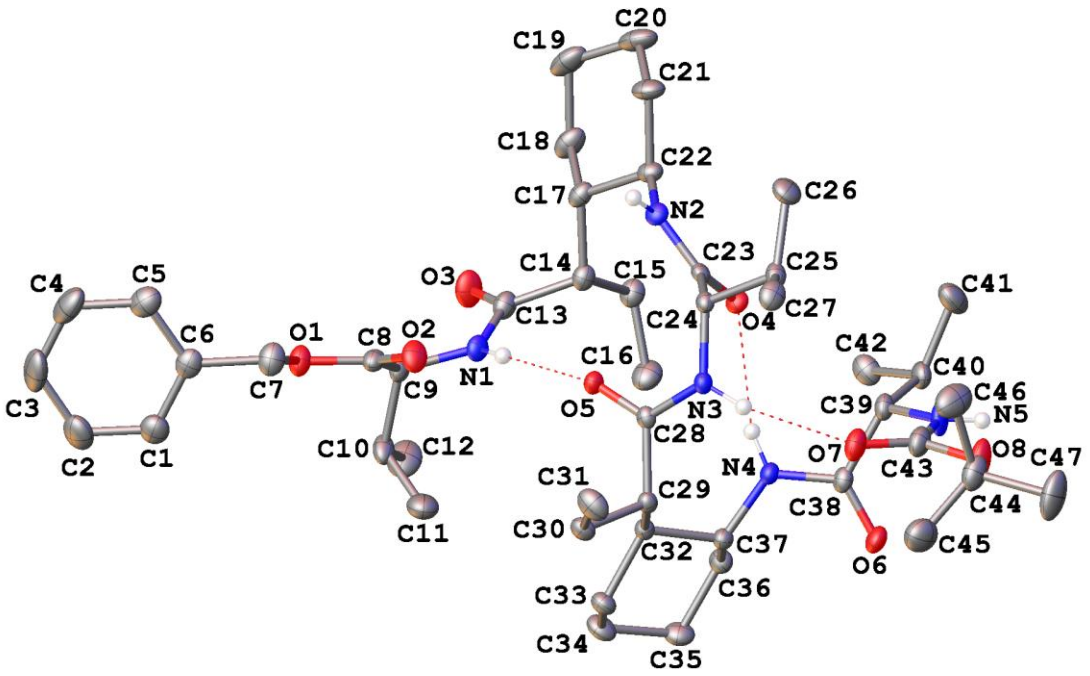


Figure S26. A molecular drawing of Gellman149 shown with 50% probability ellipsoids. All non-amide H atoms are omitted.

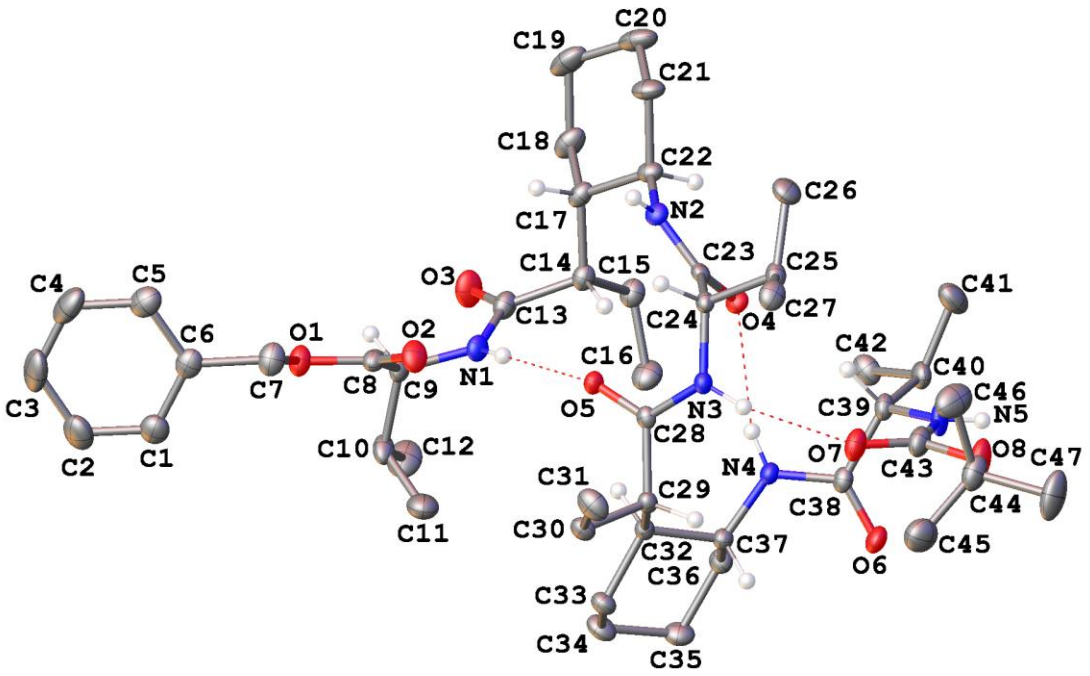


Figure S27. A molecular drawing of Gellman149 shown with 50% probability ellipsoids. All H atoms on achiral C atoms are omitted.

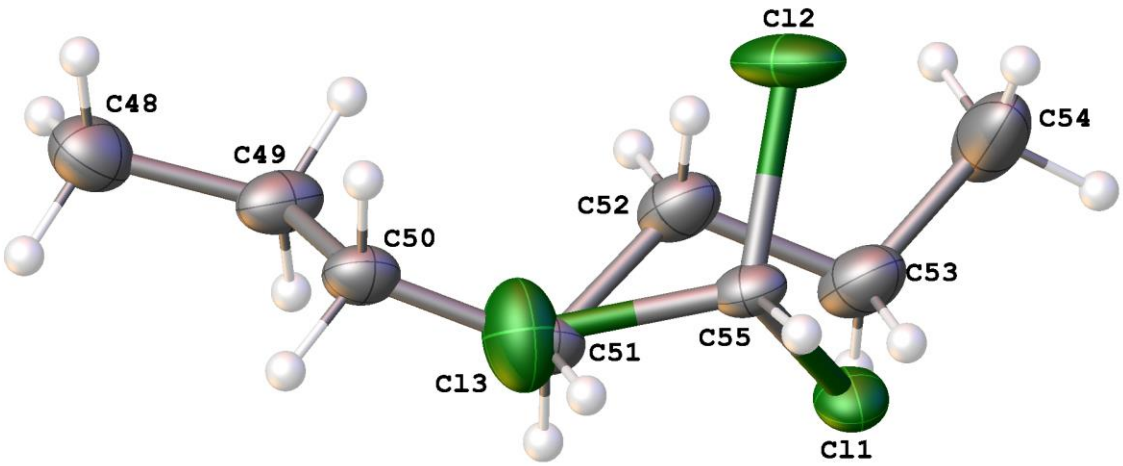


Figure S28. A molecular drawing of the solvents in Gellman149 shown with 50% probability ellipsoids. The heptane is present ~70% of the time and the chloroform ~30%.

Table S41. Crystal data and structure refinement for Gellman149	
Identification code	Gellman149
Empirical formula	$C_{47}H_{77}N_5O_8$. $(C_7H_{16})_{0.7}$. $(CHCl_3)_{0.3}$
Formula weight	946.03
Temperature/K	100.01
Crystal system	orthorhombic
Space group	$P2_12_12_1$
a/Å	12.0179(19)
b/Å	17.559(4)
c/Å	26.124(4)
α/°	90
β/°	90
γ/°	90
Volume/Å^3	5512.9(16)
Z	4
ρ_{calc}mg/mm^3	1.140
m/mm^{-1}	0.988
F(000)	2064.0
Crystal size/mm^3	0.684 × 0.524 × 0.42
2Θ range for data collection	6.064 to 146.28°
Index ranges	$-14 \le h \le 14$, $-21 \le k \le 19$, $-32 \le l \le 32$
Reflections collected	84596
Independent reflections	10935[R(int) = 0.0217]
Data/restraints/parameters	10935/6/669
Goodness-of-fit on F^2	1.097
Final R indexes [I>=2σ (I)]	R_1 = 0.0315, wR_2 = 0.0807
Final R indexes [all data]	R_1 = 0.0317, wR_2 = 0.0809
Largest diff. peak/hole / e Å^{-3}	0.42/-0.19
Flack parameter	0.02(2)

Table S42. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for Gellman149. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	*x*	*y*	*z*	**U(eq)**
O1	3311.6(11)	8846.9(7)	4760.2(5)	23.8(3)
O2	4161.3(12)	8301.1(8)	4091.1(5)	27.7(3)
O3	775.1(12)	6966.8(9)	4044.3(6)	32.5(3)
O4	3579.8(10)	5836.7(7)	2474.3(5)	18.1(2)
O5	4716.2(10)	6813.3(7)	3378.4(4)	18.0(2)
O6	5919.1(12)	3589.0(7)	2888.4(5)	24.4(3)
O7	7054.9(12)	5097.5(8)	2210.8(5)	26.8(3)
O8	7744.6(13)	4223.6(9)	1646.1(6)	37.1(4)
N1	2633.0(13)	7178.8(9)	4037.9(6)	19.6(3)
N2	3113.0(12)	7082.4(9)	2376.5(6)	17.3(3)
N3	5734.4(12)	6341.5(8)	2729.7(5)	15.7(3)
N4	5014.5(13)	4670.3(8)	3110.6(5)	16.4(3)
N5	5980.2(15)	4155.4(10)	1883.4(6)	27.0(4)

C1	4089.9(17)	9793.6(13)	5660.2(8)	29.6(4)
C2	3831.2(19)	10287.2(15)	6052.4(9)	38.4(5)
C3	3387(2)	10998.3(15)	5945.0(11)	45.4(6)
C4	3208(2)	11217.5(13)	5445.9(11)	42.3(6)
C5	3454.9(17)	10718.9(12)	5048.3(9)	31.9(5)
C6	3894.2(15)	10003.7(11)	5153.4(8)	24.6(4)
C7	4138.0(17)	9456.8(11)	4728.1(8)	28.3(4)
C8	3449.7(15)	8293(1)	4414.4(7)	19.8(4)
C9	2636.9(16)	7646.1(10)	4496.9(7)	19.9(4)
C10	2963.2(18)	7208.9(11)	4990.9(7)	26.4(4)
C11	4136(2)	6889.6(13)	4958.9(8)	34.4(5)
C12	2123(2)	6584.4(14)	5103.8(9)	42.1(6)
C13	1678.0(16)	6914.5(11)	3829.3(7)	22.8(4)
C14	1816.2(15)	6562.4(10)	3299.1(7)	19.9(3)
C15	1260.7(16)	5776.5(11)	3278.2(7)	23.5(4)
C16	1881.7(19)	5193.6(12)	3598.6(8)	31.4(4)
C17	1374.8(15)	7129.7(10)	2892.8(7)	20.5(4)
C18	97.0(16)	7118.4(11)	2853.2(9)	29.9(4)
C19	-349.7(18)	7659.8(12)	2447.8(10)	37.9(5)
C20	169.6(19)	7493.0(13)	1930.7(10)	37.7(5)
C21	1438.4(17)	7523.2(11)	1959.7(8)	28.9(4)
C22	1905.3(15)	6981.4(10)	2365.3(7)	19.7(3)
C23	3839.5(14)	6510.9(9)	2413.7(6)	15.2(3)
C24	5060.3(14)	6747.5(9)	2355.4(6)	15.1(3)
C25	5502.7(15)	6591.6(10)	1810.6(6)	19.6(4)
C26	4705.2(18)	6893.9(12)	1401.8(7)	28.7(4)
C27	6653.5(17)	6942.6(11)	1747.6(8)	26.3(4)
C28	5466.5(14)	6393.3(9)	3228.9(6)	15.1(3)
C29	6163.5(14)	5934.8(10)	3608.5(6)	15.6(3)
C30	6808.0(15)	6521.2(10)	3934.3(7)	20.3(4)
C31	7598.2(18)	6996.0(13)	3607.6(8)	30.5(4)
C32	5397.9(14)	5385.3(9)	3917.0(6)	15.9(3)
C33	5795.9(16)	5245.5(11)	4468.7(7)	20.8(4)
C34	5019.4(18)	4707.4(12)	4759.4(7)	26.5(4)
C35	4887.8(17)	3947.7(11)	4483.1(7)	25.5(4)
C36	4500.8(15)	4075.8(10)	3934.6(7)	20.7(4)
C37	5305.3(15)	4605.2(9)	3652.3(6)	17.2(3)
C38	5364.6(14)	4154.2(10)	2770.5(7)	17.9(3)
C39	5036.2(16)	4313.8(10)	2213.7(7)	20.1(4)
C40	4023.4(17)	3842.3(11)	2041.9(7)	23.7(4)
C41	3653(2)	4094.8(15)	1508.7(8)	36.3(5)
C42	3053.1(18)	3921.1(12)	2415.1(8)	27.9(4)
C43	6936.9(17)	4540.8(11)	1937.2(8)	26.2(4)
C44	8804.5(18)	4618.6(12)	1562.2(9)	31.2(5)
C45	9398(2)	4775.1(16)	2062.2(10)	42.1(6)
C46	8578(2)	5336.6(15)	1259.4(10)	43.7(6)
C47	9452(2)	4045.8(15)	1241.0(12)	49.3(7)
C48	3223(3)	4289(2)	6266.7(15)	44.0(8)
C49	2926(3)	3958(2)	5760.8(12)	35.9(7)
C50	1982(3)	4380.8(17)	5489.1(11)	30.9(6)
C51	1771(8)	4107(4)	4950(3)	30.9(6)

C52	1422(3)	3274.1(19)	4913.2(13)	38.2(8)
C53	1305(4)	2981(2)	4364.9(13)	43.9(9)
C54	909(5)	2157(2)	4350.6(15)	61.2(13)
Cl1	1976.4(14)	3169.6(10)	4142.7(6)	32.4(5)
Cl2	540(2)	2731.2(11)	4971.6(8)	49.9(7)
Cl3	1786(6)	4150(3)	5024.0(19)	56.1(11)
C55	1074(6)	3507(4)	4618(3)	26.2(14)

Table S43. Anisotropic Displacement Parameters ($Å^2×10^3$) for Gellman149. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+...+2hka×b×U_{12}]$

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O1	26.0(7)	19.6(6)	25.9(6)	-4.3(5)	7.8(5)	-2.7(5)
O2	35.4(8)	25.6(7)	22.1(6)	0.6(5)	11.9(6)	-3.2(6)
O3	22.3(7)	38.9(8)	36.4(8)	-9.7(7)	12.1(6)	-3.6(6)
O4	21.0(6)	12.8(6)	20.6(6)	1.1(5)	0.2(5)	-1.1(5)
O5	18.5(6)	18.3(6)	17.2(6)	-2.8(5)	1.6(5)	3.3(5)
O6	29.4(7)	16.6(6)	27.1(6)	-3.7(5)	1.8(5)	5.5(5)
O7	29.2(7)	21.7(7)	29.4(7)	-5.8(5)	10.2(6)	1.5(5)
O8	35.6(8)	29.3(8)	46.4(9)	-16.1(7)	21.4(7)	-3.1(6)
N1	19.4(7)	20.6(7)	18.8(7)	-1.0(6)	7.3(6)	0.8(6)
N2	19.6(7)	12.4(7)	20.0(7)	2.2(5)	0.5(6)	-2.9(6)
N3	17.3(7)	15.1(7)	14.6(7)	-2.3(5)	1.2(5)	0.7(6)
N4	19.4(7)	13.7(7)	16.3(7)	-1.1(5)	1.2(6)	2.0(6)
N5	33.1(9)	25.4(8)	22.5(8)	-10.9(7)	10.2(7)	0.6(7)
C1	22.7(9)	31.9(10)	34.1(11)	-3.7(8)	-4.5(8)	-2.3(8)
C2	31.3(11)	48.0(13)	36.0(11)	-13.2(10)	-8.8(9)	-4.6(10)
C3	37.5(12)	42.1(13)	56.5(15)	-31.2(12)	-4.9(11)	-2.6(10)
C4	34.4(12)	22(1)	70.5(17)	-12.0(11)	-7.1(11)	2.6(9)
C5	27.4(10)	25(1)	43.2(12)	-0.3(9)	-3.4(9)	-4.4(8)
C6	17.2(8)	23.3(9)	33.4(10)	-2.8(8)	1.5(7)	-5.2(7)
C7	26.5(10)	24.5(9)	33.8(10)	-2.6(8)	7.9(8)	-5.8(8)
C8	24.6(9)	17.8(8)	17.2(8)	2.5(6)	2.1(7)	3.1(7)
C9	23.2(9)	17.5(8)	18.9(8)	-1.1(7)	7.1(7)	1.4(7)
C10	42.9(11)	19.2(9)	17.0(8)	0.0(7)	9.2(8)	0.1(8)
C11	43.3(12)	33.1(11)	26.7(10)	6.1(9)	-4.6(9)	5.1(10)
C12	56.6(15)	31.2(11)	38.5(12)	12.3(9)	17.8(11)	-3.8(11)
C13	22.9(9)	19.8(9)	25.7(9)	-0.5(7)	6.7(7)	0.3(7)
C14	17.1(8)	18.8(8)	23.9(9)	-1.8(7)	3.0(7)	-1.4(7)
C15	24.1(9)	19.7(9)	26.9(9)	-0.1(7)	4.5(7)	-3.5(7)
C16	38.1(12)	22.4(9)	33.8(11)	6.2(8)	5.7(9)	-0.3(9)
C17	16.7(8)	14.9(8)	29.8(9)	-0.4(7)	1.2(7)	-0.4(7)
C18	17.3(9)	23.9(9)	48.6(12)	-1.4(9)	0.7(8)	0.2(7)
C19	21.1(9)	25.5(10)	67.1(16)	0.1(10)	-10.2(10)	4.1(8)
C20	31.4(11)	28.5(11)	53.1(14)	10(1)	-19.7(10)	1.4(9)
C21	30.4(10)	23.6(9)	32.6(10)	9.6(8)	-10.0(8)	1.3(8)
C22	20.5(9)	15.4(8)	23.3(8)	3.4(7)	-4.3(7)	-0.8(7)
C23	19.8(8)	15.9(8)	10.0(7)	0.2(6)	-0.4(6)	-0.8(6)
C24	20.0(8)	12.3(7)	13.1(7)	-0.4(6)	1.3(6)	-0.4(6)

C25	27.1(9)	16.5(8)	15.1(8)	-0.5(6)	4.8(7)	-1.4(7)
C26	35.5(11)	33.6(11)	17.0(8)	4.1(8)	1.6(8)	0.5(9)
C27	28.8(10)	25.3(9)	24.8(9)	2.7(8)	10.1(8)	-1.2(8)
C28	15.5(8)	14.3(8)	15.4(7)	-1.7(6)	0.9(6)	-3.4(6)
C29	14.3(7)	17.4(8)	15.0(7)	-2.0(6)	0.5(6)	0.2(6)
C30	19.6(8)	23.5(9)	18.0(8)	-0.3(7)	-2.8(7)	-2.4(7)
C31	29.9(10)	35.1(11)	26.4(9)	-0.1(8)	-2.0(8)	-13.9(9)
C32	17.2(8)	16.2(8)	14.4(7)	-0.9(6)	0.9(6)	0.8(6)
C33	23.1(9)	23.5(9)	15.8(8)	1.3(7)	-2.3(7)	-1.5(7)
C34	32.5(10)	31.7(10)	15.4(8)	5.4(7)	0.2(7)	-5.9(8)
C35	28.4(10)	25.3(10)	22.8(9)	9.3(7)	-0.2(8)	-2.6(8)
C36	24.9(9)	17.4(8)	19.9(8)	0.4(7)	0.0(7)	-2.5(7)
C37	19.1(8)	16.7(8)	15.8(8)	-0.7(6)	0.5(6)	2.4(7)
C38	18.6(8)	14.0(8)	21.1(8)	-2.7(6)	3.0(7)	-2.8(7)
C39	25.4(9)	16.2(8)	18.7(8)	-3.3(6)	5.3(7)	0.2(7)
C40	30.4(10)	18.7(8)	22.0(9)	-6.5(7)	-1.2(7)	0.0(7)
C41	34.2(11)	53.7(14)	21.1(10)	-5.5(9)	-1.3(8)	3.8(10)
C42	30.1(10)	27.7(10)	25.8(9)	-0.6(8)	0.0(8)	-10.9(8)
C43	31.2(10)	22.5(9)	25.0(9)	-2.4(8)	10.0(8)	3.7(8)
C44	31.0(11)	25.4(10)	37.2(11)	-4.3(8)	13.9(9)	0.6(8)
C45	36.3(12)	48.7(14)	41.3(13)	-0.9(11)	5.5(10)	10.6(10)
C46	43.8(13)	46.7(14)	40.7(12)	11.7(11)	17.3(11)	8.9(11)
C47	40.7(13)	38.9(13)	68.3(17)	-21.0(12)	30.4(13)	-3.3(11)
C48	39.3(18)	44.3(19)	48(2)	7.1(16)	-3.2(16)	-9.3(15)
C49	29.1(15)	44.0(18)	34.5(16)	13.3(14)	1.7(13)	0.2(13)
C50	32.3(14)	28.6(13)	31.8(14)	9.0(11)	0.2(11)	0.2(11)
C51	32.3(14)	28.6(13)	31.8(14)	9.0(11)	0.2(11)	0.2(11)
C52	48.7(19)	36.8(17)	29.1(16)	7.1(14)	1.2(15)	4.8(15)
C53	61(2)	40.4(19)	30.3(16)	6.6(15)	4.0(17)	4.9(17)
C54	114(4)	38(2)	31.7(18)	-1.0(15)	-12(2)	-1(2)
Cl1	28.7(9)	37.8(9)	30.9(9)	-0.9(7)	9.6(7)	-5.2(7)
Cl2	64.3(14)	34.8(10)	50.7(12)	17.0(9)	29.8(10)	4.1(9)
Cl3	51.2(16)	56.6(17)	60(2)	-22.1(15)	-6.2(17)	2.3(13)
C55	29(3)	27(3)	22(3)	5(3)	7(3)	3(3)

Table S44. Bond Lengths for Gellman149.

Atom	**Atom**	**Length/Å**	**Atom**	**Atom**	**Length/Å**
O1	C7	1.463(2)	C17	C22	1.540(3)
O1	C8	1.338(2)	C18	C19	1.521(3)
O2	C8	1.202(2)	C19	C20	1.517(4)
O3	C13	1.225(2)	C20	C21	1.528(3)
O4	C23	1.234(2)	C21	C22	1.531(2)
O5	C28	1.229(2)	C23	C24	1.532(2)
O6	C38	1.234(2)	C24	C25	1.544(2)
O7	C43	1.219(2)	C25	C26	1.530(3)
O8	C43	1.353(2)	C25	C27	1.523(3)
O8	C44	1.467(3)	C28	C29	1.528(2)
N1	C9	1.453(2)	C29	C30	1.544(2)

N1	C13	1.353(3)	C29	C32	1.558(2)
N2	C22	1.463(2)	C30	C31	1.525(3)
N2	C23	1.334(2)	C32	C33	1.538(2)
N3	C24	1.456(2)	C32	C37	1.538(2)
N3	C28	1.346(2)	C33	C34	1.530(3)
N4	C37	1.462(2)	C34	C35	1.525(3)
N4	C38	1.337(2)	C35	C36	1.523(3)
N5	C39	1.452(2)	C36	C37	1.531(2)
N5	C43	1.341(3)	C38	C39	1.533(2)
C1	C2	1.378(3)	C39	C40	1.539(3)
C1	C6	1.394(3)	C40	C41	1.528(3)
C2	C3	1.387(4)	C40	C42	1.526(3)
C3	C4	1.376(4)	C44	C45	1.514(3)
C4	C5	1.390(3)	C44	C46	1.513(3)
C5	C6	1.390(3)	C44	C47	1.523(3)
C6	C7	1.497(3)	C48	C49	1.488(5)
C8	C9	1.514(3)	C49	C50	1.530(4)
C9	C10	1.552(3)	C50	C51	1.510(7)
C10	C11	1.519(3)	C51	C52	1.525(7)
C10	C12	1.520(3)	C52	C53	1.528(4)
C13	C14	1.526(2)	C53	C54	1.524(5)
C14	C15	1.534(2)	Cl1	C55	1.752(7)
C14	C17	1.549(3)	Cl2	C55	1.766(7)
C15	C16	1.518(3)	Cl3	C55	1.770(9)
C17	C18	1.539(2)			

Table S45. Bond Angles for Gellman149.

Atom	**Atom**	**Atom**	**Angle/°**	**Atom**	**Atom**	**Atom**	**Angle/°**
C8	O1	C7	114.17(14)	C23	C24	C25	111.92(14)
C43	O8	C44	120.82(16)	C26	C25	C24	111.49(15)
C13	N1	C9	121.94(15)	C27	C25	C24	109.91(14)
C23	N2	C22	124.05(15)	C27	C25	C26	110.67(15)
C28	N3	C24	118.97(14)	O5	C28	N3	121.60(16)
C38	N4	C37	120.99(15)	O5	C28	C29	120.82(15)
C43	N5	C39	120.71(16)	N3	C28	C29	117.52(15)
C2	C1	C6	120.1(2)	C28	C29	C30	106.35(14)
C1	C2	C3	120.2(2)	C28	C29	C32	109.79(13)
C4	C3	C2	120.2(2)	C30	C29	C32	115.09(14)
C3	C4	C5	119.9(2)	C31	C30	C29	111.62(15)
C6	C5	C4	120.2(2)	C33	C32	C29	113.58(14)
C1	C6	C7	120.12(19)	C33	C32	C37	107.55(14)
C5	C6	C1	119.4(2)	C37	C32	C29	111.20(13)
C5	C6	C7	120.5(2)	C34	C33	C32	111.96(15)
O1	C7	C6	107.10(15)	C35	C34	C33	111.63(16)
O1	C8	C9	111.69(15)	C36	C35	C34	110.35(15)
O2	C8	O1	123.66(17)	C35	C36	C37	110.49(15)
O2	C8	C9	124.61(17)	N4	C37	C32	112.51(14)
N1	C9	C8	107.97(14)	N4	C37	C36	111.27(14)

N1	C9	C10	114.06(15)	C36	C37	C32	111.71(14)
C8	C9	C10	109.06(15)	O6	C38	N4	123.30(17)
C11	C10	C9	111.79(16)	O6	C38	C39	121.52(15)
C11	C10	C12	111.16(18)	N4	C38	C39	115.18(15)
C12	C10	C9	110.51(18)	N5	C39	C38	109.12(15)
O3	C13	N1	122.75(17)	N5	C39	C40	109.97(15)
O3	C13	C14	122.88(18)	C38	C39	C40	112.45(15)
N1	C13	C14	114.35(16)	C41	C40	C39	109.89(17)
C13	C14	C15	110.45(15)	C42	C40	C39	111.67(15)
C13	C14	C17	108.92(15)	C42	C40	C41	109.46(17)
C15	C14	C17	113.90(15)	O7	C43	O8	125.18(19)
C16	C15	C14	111.90(16)	O7	C43	N5	124.44(18)
C18	C17	C14	112.29(15)	N5	C43	O8	110.38(17)
C18	C17	C22	110.53(16)	O8	C44	C45	111.48(18)
C22	C17	C14	111.25(14)	O8	C44	C46	108.41(19)
C19	C18	C17	113.00(17)	O8	C44	C47	102.32(17)
C20	C19	C18	110.74(18)	C45	C44	C47	110.8(2)
C19	C20	C21	111.10(19)	C46	C44	C45	112.6(2)
C20	C21	C22	112.24(17)	C46	C44	C47	110.7(2)
N2	C22	C17	111.86(14)	C48	C49	C50	113.6(3)
N2	C22	C21	107.59(15)	C51	C50	C49	113.8(4)
C21	C22	C17	111.26(16)	C50	C51	C52	114.2(5)
O4	C23	N2	124.43(16)	C51	C52	C53	114.0(4)
O4	C23	C24	120.98(15)	C54	C53	C52	111.8(3)
N2	C23	C24	114.55(14)	Cl1	C55	Cl2	109.6(4)
N3	C24	C23	109.46(13)	Cl1	C55	Cl3	109.9(4)
N3	C24	C25	109.92(14)	Cl2	C55	Cl3	110.7(4)

Table S46. Hydrogen Bonds for Gellman149.

D	**H**	**A**	**d(D-H)/Å**	**d(H-A)/Å**	**d(D-A)/Å**	**D-H-A/°**
N1	H1	O5	0.87(2)	2.26(2)	3.1061(19)	164(2)
N2	H2	O6[1]	0.85(2)	2.13(3)	2.972(2)	170(2)
N3	H3	O7	0.87(3)	2.17(3)	3.021(2)	168(2)
N4	H4	O4	0.82(3)	2.35(3)	3.151(2)	163(2)
N5	H5	O2[2]	0.79(3)	2.26(3)	2.960(2)	148(2)

[1]1-X,1/2+Y,1/2-Z; [2]1-X,-1/2+Y,1/2-Z

Table S47. Torsion Angles for Gellman149.

A	**B**	**C**	**D**	**Angle/°**	**A**	**B**	**C**	**D**	**Angle/°**
O1	C8	C9	N1	164.93(15)	C17	C18	C19	C20	55.6(2)
O1	C8	C9	C10	-70.63(19)	C18	C17	C22	N2	172.87(14)
O2	C8	C9	N1	-17.6(3)	C18	C17	C22	C21	52.5(2)
O2	C8	C9	C10	106.8(2)	C18	C19	C20	C21	-55.6(2)
O3	C13	C14	C15	-51.4(2)	C19	C20	C21	C22	56.1(2)
O3	C13	C14	C17	74.4(2)	C20	C21	C22	N2	-177.42(17)

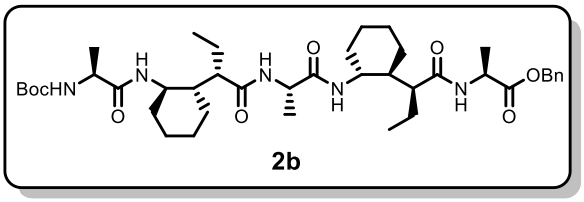
O4	C23	C24	N3	-43.5(2)	C20	C21	C22	C17	-54.6(2)
O4	C23	C24	C25	78.62(19)	C22	N2	C23	O4	-4.6(3)
O5	C28	C29	C30	-64.75(19)	C22	N2	C23	C24	173.22(15)
O5	C28	C29	C32	60.4(2)	C22	C17	C18	C19	-53.9(2)
O6	C38	C39	N5	-42.0(2)	C23	N2	C22	C17	99.32(19)
O6	C38	C39	C40	80.3(2)	C23	N2	C22	C21	-138.20(17)
N1	C9	C10	C11	61.7(2)	C23	C24	C25	C26	48.42(19)
N1	C9	C10	C12	-62.7(2)	C23	C24	C25	C27	171.51(14)
N1	C13	C14	C15	129.96(17)	C24	N3	C28	O5	-4.9(2)
N1	C13	C14	C17	-104.25(18)	C24	N3	C28	C29	177.63(14)
N2	C23	C24	N3	138.56(14)	C28	N3	C24	C23	-55.42(19)
N2	C23	C24	C25	-99.32(17)	C28	N3	C24	C25	-178.72(14)
N3	C24	C25	C26	170.26(15)	C28	C29	C30	C31	-62.36(19)
N3	C24	C25	C27	-66.64(18)	C28	C29	C32	C33	-147.52(15)
N3	C28	C29	C30	112.74(16)	C28	C29	C32	C37	91.00(16)
N3	C28	C29	C32	-122.15(16)	C29	C32	C33	C34	179.67(15)
N4	C38	C39	N5	137.57(16)	C29	C32	C37	N4	-50.62(19)
N4	C38	C39	C40	-100.13(18)	C29	C32	C37	C36	-176.61(14)
N5	C39	C40	C41	-64.9(2)	C30	C29	C32	C33	-27.6(2)
N5	C39	C40	C42	173.39(16)	C30	C29	C32	C37	-149.08(15)
C1	C2	C3	C4	-0.5(4)	C32	C29	C30	C31	175.84(16)
C1	C6	C7	O1	70.1(2)	C32	C33	C34	C35	56.7(2)
C2	C1	C6	C5	1.1(3)	C33	C32	C37	N4	-175.56(14)
C2	C1	C6	C7	-177.85(19)	C33	C32	C37	C36	58.45(18)
C2	C3	C4	C5	1.2(4)	C33	C34	C35	C36	-55.0(2)
C3	C4	C5	C6	-0.8(3)	C34	C35	C36	C37	56.0(2)
C4	C5	C6	C1	-0.3(3)	C35	C36	C37	N4	173.93(15)
C4	C5	C6	C7	178.61(19)	C35	C36	C37	C32	-59.41(19)
C5	C6	C7	O1	-108.8(2)	C37	N4	C38	O6	2.0(3)
C6	C1	C2	C3	-0.7(3)	C37	N4	C38	C39	-177.57(15)
C7	O1	C8	O2	-2.6(3)	C37	C32	C33	C34	-56.84(19)
C7	O1	C8	C9	174.86(15)	C38	N4	C37	C32	149.49(15)
C8	O1	C7	C6	-177.41(16)	C38	N4	C37	C36	-84.28(19)
C8	C9	C10	C11	-59.1(2)	C38	C39	C40	C41	173.24(16)
C8	C9	C10	C12	176.53(16)	C38	C39	C40	C42	51.6(2)
C9	N1	C13	O3	-9.5(3)	C39	N5	C43	O7	-9.2(3)
C9	N1	C13	C14	169.20(15)	C39	N5	C43	O8	170.71(17)
C13	N1	C9	C8	-136.40(17)	C43	O8	C44	C45	59.2(3)
C13	N1	C9	C10	102.2(2)	C43	O8	C44	C46	-65.3(3)
C13	C14	C15	C16	-69.5(2)	C43	O8	C44	C47	177.6(2)
C13	C14	C17	C18	-79.11(19)	C43	N5	C39	C38	-60.9(2)
C13	C14	C17	C22	156.42(15)	C43	N5	C39	C40	175.31(17)
C14	C17	C18	C19	-178.76(17)	C44	O8	C43	O7	-11.6(3)
C14	C17	C22	N2	-61.67(19)	C44	O8	C43	N5	168.43(18)
C14	C17	C22	C21	177.96(15)	C48	C49	C50	C51	173.0(4)
C15	C14	C17	C18	44.7(2)	C49	C50	C51	C52	61.6(7)
C15	C14	C17	C22	-79.82(18)	C50	C51	C52	C53	-175.1(5)
C17	C14	C15	C16	167.55(16)	C51	C52	C53	C54	-177.6(5)

Table S48. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for Gellman149.

Atom	*x*	*y*	*z*	**U(eq)**
H1	3250(20)	7171(13)	3866(9)	22(5)
H2	3362(19)	7530(14)	2336(8)	20(5)
H3	6170(20)	5985(15)	2622(10)	31(6)
H4	4630(20)	5025(14)	3002(9)	24(6)
H5	5980(20)	3789(16)	1707(10)	31(6)
H1A	4402	9309	5735	35
H2A	3958	10140	6397	46
H3A	3204	11335	6217	54
H4A	2917	11709	5373	51
H5A	3323	10867	4704	38
H7A	4086	9718	4393	34
H7B	4898	9246	4765	34
H9	1876	7866	4545	24
H10	2943	7579	5282	32
H11A	4667	7309	4919	52
H11B	4192	6547	4664	52
H11C	4305	6607	5273	52
H12A	2136	6207	4827	63
H12B	1377	6807	5129	63
H12C	2313	6336	5428	63
H14	2630	6490	3237	24
H15A	486	5819	3404	28
H15B	1233	5600	2918	28
H16A	1900	5362	3956	47
H16B	2644	5142	3470	47
H16C	1502	4701	3576	47
H17	1600	7653	3003	25
H18A	-150	6594	2772	36
H18B	-223	7258	3189	36
H19A	-1168	7605	2424	46
H19B	-182	8191	2548	46
H20A	-97	7871	1678	45
H20B	-66	6982	1814	45
H21A	1674	8050	2041	35
H21B	1753	7387	1622	35
H22	1737	6446	2260	24
H24	5121	7306	2425	18
H25	5571	6028	1765	23
H26A	4555	7434	1465	43
H26B	4006	6607	1416	43
H26C	5044	6834	1063	43
H27A	7159	6729	2005	39
H27B	6605	7496	1793	39
H27C	6938	6829	1404	39
H29	6713	5623	3412	19
H30A	6272	6863	4108	24
H30B	7239	6250	4201	24
H31A	8051	7322	3830	46

H31B	7166	7314	3372	46
H31C	8085	6657	3411	46
H32	4637	5615	3932	19
H33A	6554	5024	4461	25
H33B	5836	5738	4652	25
H34A	5322	4616	5107	32
H34B	4280	4950	4797	32
H35A	5608	3674	4482	31
H35B	4338	3629	4667	31
H36A	3747	4304	3936	25
H36B	4459	3581	3754	25
H37	6058	4364	3669	21
H39	4845	4866	2182	24
H40	4246	3294	2026	28
H41A	4260	4012	1264	54
H41B	3459	4637	1516	54
H41C	3001	3798	1404	54
H42A	2838	4458	2441	42
H42B	3278	3733	2753	42
H42C	2419	3623	2291	42
H45A	9372	4319	2278	63
H45B	9032	5199	2239	63
H45C	10175	4909	1993	63
H46A	8124	5685	1465	66
H46B	8178	5207	944	66
H46C	9284	5583	1173	66
H47A	9044	3943	924	74
H47B	9541	3571	1434	74
H47C	10186	4255	1159	74
H48A	3411	4829	6224	66
H48B	3865	4017	6408	66
H48C	2590	4241	6501	66
H49A	2704	3420	5810	43
H49B	3593	3964	5538	43
H50A	1291	4320	5691	37
H50B	2163	4931	5479	37
H51A	2457	4181	4746	37
H51B	1181	4426	4795	37
H52A	701	3209	5091	46
H52B	1980	2958	5094	46
H53A	768	3305	4178	53
H53B	2033	3020	4189	53
H54A	1358	1851	4587	92
H54B	990	1956	4003	92
H54C	126	2133	4453	92
H55	441	3780	4450	31

Crystallographic Experimental Section for Compound 2b



2b

Data Collection

A colorless crystal with approximate dimensions 0.28 x 0.24 x 0.11 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker Venture MX diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from a series of ω scans with the exposure time of 20 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9777 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.80 Å. A total of 45447 data were harvested by collecting 12 sets of frames with 0.2º scans in ω and φ with an exposure time 60 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were consistent for the space groups $P2_1$ and $P2_1/m$. The *E*-statistics strongly suggested the non-centrosymmetric space group $P2_1$ that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All polypeptide non-hydrogen atoms were refined with anisotropic displacement coefficients. All hydrogen atoms were included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

There are two symmetry independent polypeptide molecules in the asymmetric unit. The molecular conformations differ slightly, most noticeably in the position of the tolyl group.

There are also four disordered solvent molecules in the unit cell: two heptanes and two ethanols. The heptanes are equally disordered over two positions each. Ethanol O9 is disordered over two positions (major component occupancy is 73.2(16)%), in ethanol O10 only the ethyl groups are disordered with the major component occupied 68.5(19)% of the time. In the solvents the O9/O9a molecules and atom O10 were refined anisotropically, all other atoms isotropically. Sixty three restraints and constraints were applied to the solvent molecules to ensure computationally stable and chemically reasonable refinement.

The proposed absolute configuration was confirmed experimentally by the Hooft y parameter (0.08(6)), but it was inconclusive according to the Flack x parameter (0.1(2)).

The final least-squares refinement of 1127 parameters against 20128 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0894 and 0.2606, respectively. The final difference Fourier map was featureless.

Summary

Crystal Data for $C_{50}H_{87}N_5O_9$ (*M* =902.24): monoclinic, space group $P2_1$ (no. 4), *a* = 13.4361(18) Å, *b* = 18.178(3) Å, *c* = 22.458(3) Å, *β* = 100.534(9)°, *V* = 5392.7(13) $Å^3$,*Z* = 4, *T* = 100.15 K, μ(CuKα) = 0.605 mm^{-1}, *Dcalc* = 1.111 g/mm^3, 45447 reflections measured ($6.298 \leq 2\Theta \leq 149.016$), 20128 unique ($R_{int}$ = 0.0386, R_{sigma} = 0.0485) which were used in all calculations. The final R_1 was 0.0894 ($I > 2\sigma(I)$) and wR_2 was 0.2606 (all data).

References

[1] Bruker-AXS. (2007-2013) APEX2 (Ver. 2013.2-0), SADABS (2012-1), and SAINT+ (Ver. 8.30C) Software Reference Manuals. Bruker-AXS, Madison, Wisconsin, USA.

[2] Sheldrick, G. M. (2008) SHELXL. *Acta Cryst.* **A64**, 112-122.

[3] Dolomanov, O.V.; Bourhis, L.J.; Gildea, R.J.; Howard, J.A.K.; Puschmann, H. "OLEX2: a complete structure solution, refinement and analysis program". *J. Appl. Cryst.* (2009) **42**, *339-341*.

[4] Guzei, I.A. (2013). Internal laboratory computer programs Gn.

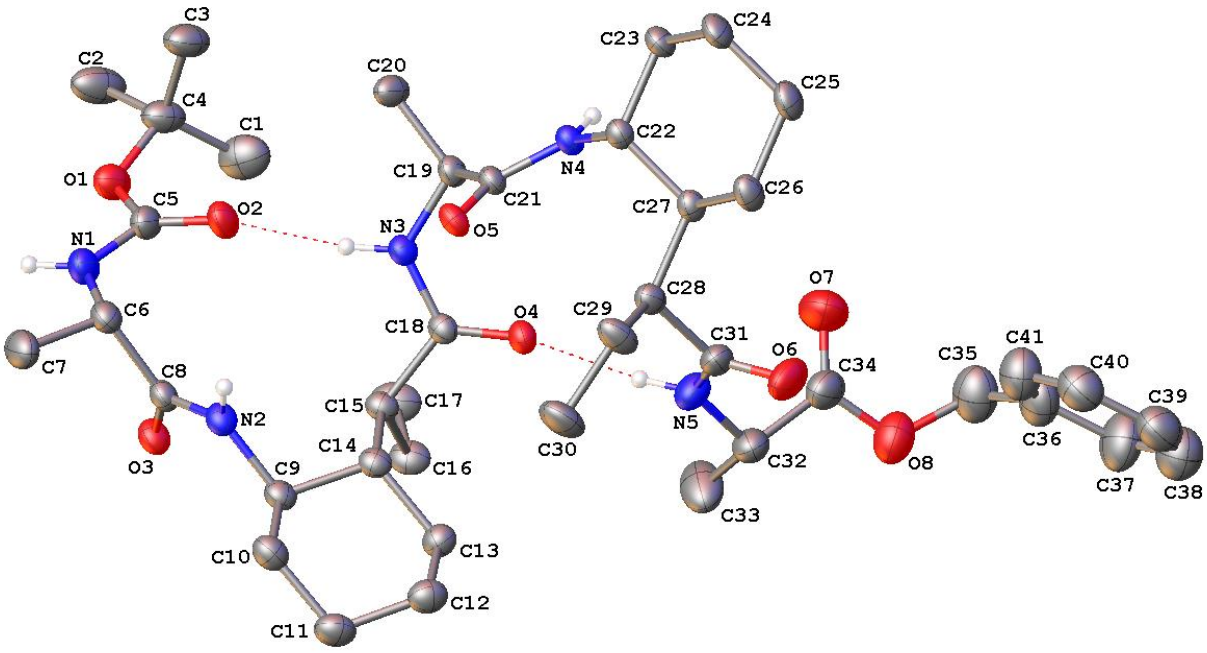


Figure S29. A molecular drawing of the O1 foldamer shown with 40% probability ellipsoids. All aliphatic and aromatic H atoms are omitted.

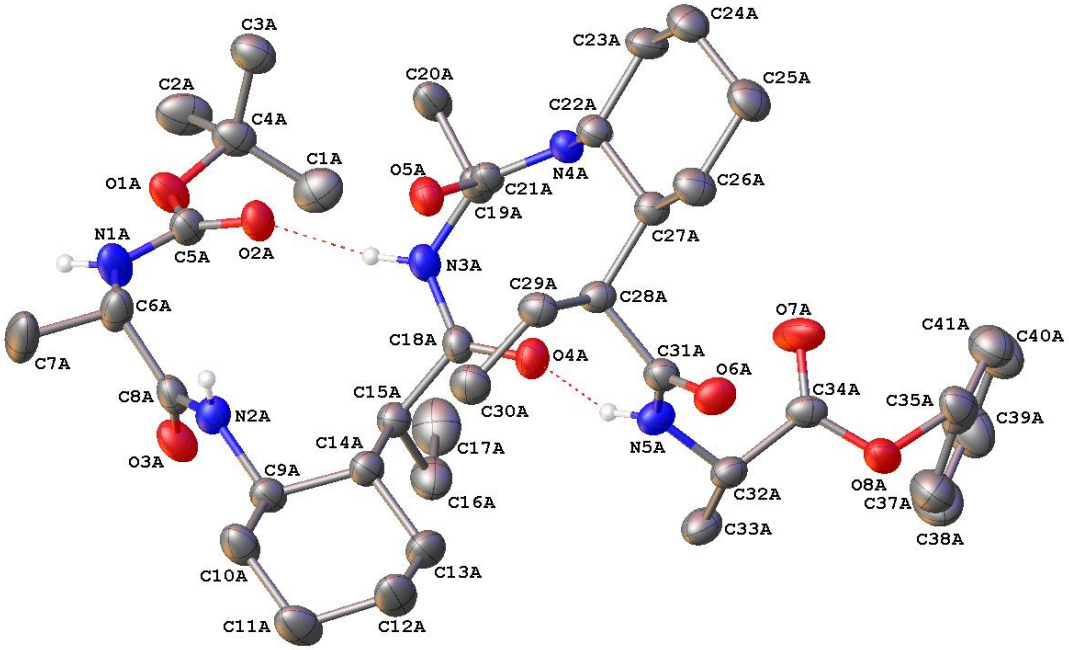


Figure S30. A molecular drawing of the O1a foldamer shown with 40% probability ellipsoids. All aliphatic and aromatic H atoms are omitted.

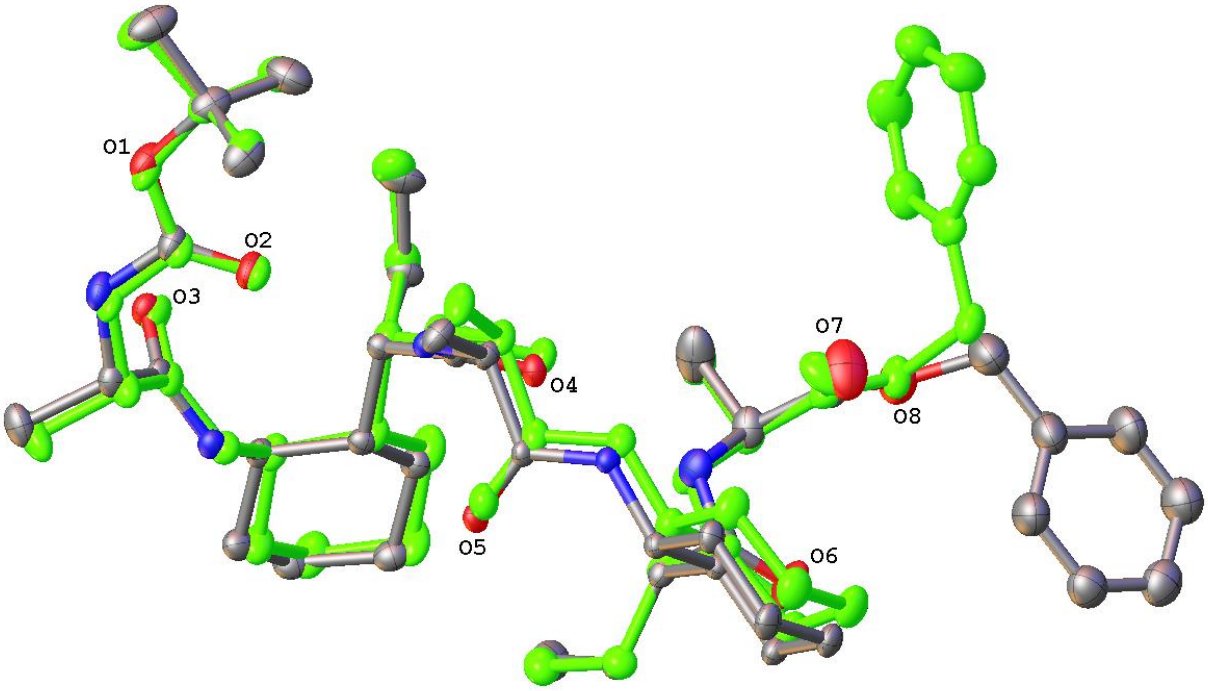


Figure S31. Superposition of the two polypeptides shown with 30% probability ellipsoids. All H atoms are omitted.

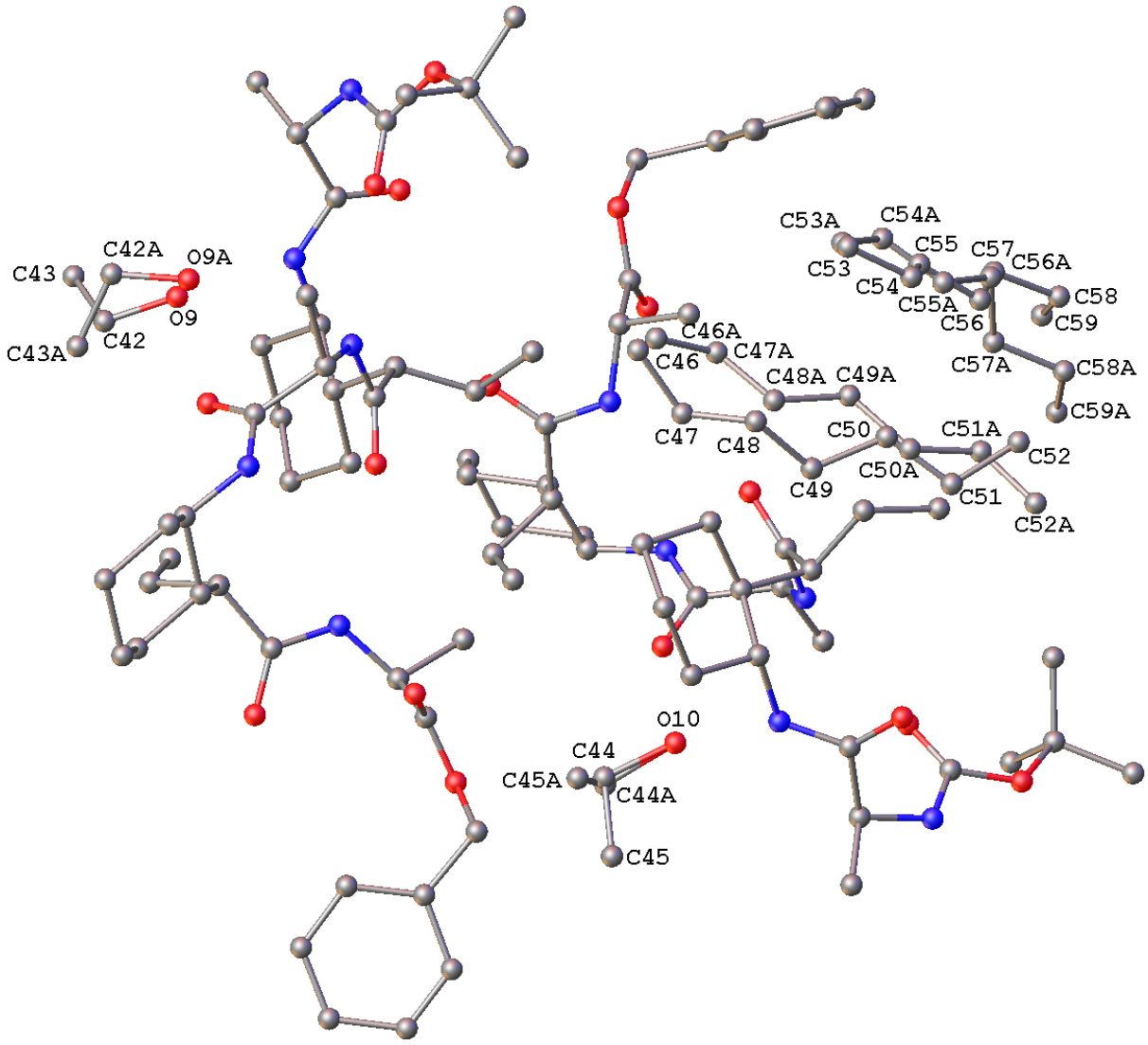


Figure S32. Content of the asymmetric unit. All H atoms are omitted, but all molecules with disordered atoms are shown. Only the solvent molecules are labeled.

Table S49. Crystal data and structure refinement for Gellman153.

Identification code	Gellman153
Empirical formula	$C_{41}H_{65}N_5O_8$ .C_7H_{16} . EtOH
Formula weight	902.24
Temperature/K	100.15
Crystal system	monoclinic
Space group	$P2_1$
a/Å	13.4361(18)
b/Å	18.178(3)
c/Å	22.458(3)
α/°	90
β/°	100.534(9)
γ/°	90
Volume/Å^3	5392.7(13)
Z	4
ρ_{calc}mg/mm^3	1.111
m/mm^{-1}	0.605
F(000)	1976.0
Crystal size/mm^3	0.28 × 0.24 × 0.11
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection	6.298 to 149.016°
Index ranges	$-15 \leq h \leq 16$, $-22 \leq k \leq 19$, $-25 \leq l \leq 28$
Reflections collected	45447
Independent reflections	20128 [R_{int} = 0.0386, R_{sigma} = 0.0485]
Data/restraints/parameters	20128/64/1127
Goodness-of-fit on F^2	0.967
Final R indexes [I>=2σ (I)]	R_1 = 0.0894, wR_2 = 0.2495
Final R indexes [all data]	R_1 = 0.0939, wR_2 = 0.2606
Largest diff. peak/hole / e Å^{-3}	0.66/-0.51
Flack parameter	0.1(2)
Hooft parameter	0.08(6)

Table S50. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for Gellman153. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	*x*	*y*	*z*	U(eq)
O1	7821(2)	1462(2)	6010.6(16)	48.4(7)
O2	6177(2)	1117.9(19)	5673.7(16)	46.0(7)
O3	5630(2)	2760.4(17)	4907.8(13)	38.9(6)
O4	3457(2)	23.6(16)	4115.6(12)	35.3(5)
O5	2759(2)	-0.6(15)	5457.6(12)	32.8(5)
O6	68(3)	-1120(3)	3435.3(18)	66.1(11)
O7	2010(6)	-1980(3)	3026(3)	97.2(19)
O8	1227(4)	-1716(3)	2093(2)	75.4(12)
N1	6578(3)	2255(2)	6053.3(19)	46.5(8)
N2	4111(2)	2419.5(19)	5120.6(14)	33.6(6)

N3	4519(2)	259.9(18)	4993.9(14)	29.8(6)
N4	2896(2)	-1214.8(17)	5242.5(13)	28.1(6)
N5	1580(3)	-611(3)	3416.1(17)	48.6(9)
C1	7989(5)	403(4)	5355(3)	68.1(15)
C2	9371(4)	858(4)	6189(3)	69.4(17)
C3	7842(4)	234(4)	6454(3)	60.4(13)
C4	8236(4)	719(3)	5988(2)	51.0(11)
C5	6806(3)	1574(3)	5895.7(19)	40.1(8)
C6	5532(3)	2500(2)	5951.0(18)	38.3(8)
C7	5463(5)	3251(3)	6254(2)	53.5(11)
C8	5102(3)	2561(2)	5274.2(17)	32.5(7)
C9	3541(3)	2475(2)	4498.7(16)	32.6(7)
C10	2619(3)	2965(2)	4498(2)	41.2(8)
C11	1938(3)	3004(3)	3872(2)	45.8(9)
C12	1613(3)	2238(3)	3653(2)	43.2(9)
C13	2532(3)	1748(2)	3634.8(17)	37.6(8)
C14	3247(3)	1700(2)	4253.6(15)	31.2(7)
C15	4216(3)	1235(2)	4248.8(16)	31.0(7)
C16	4613(3)	1248(3)	3646.4(18)	40.9(8)
C17	5673(4)	924(4)	3726(3)	57.2(13)
C18	4035(3)	450(2)	4437.8(15)	29.1(6)
C19	4364(3)	-454(2)	5249.4(16)	31.3(7)
C20	5050(3)	-537(3)	5870.1(19)	41.7(9)
C21	3256(2)	-532.4(19)	5321.3(14)	27.1(6)
C22	1889(3)	-1453(2)	5321.9(16)	29.1(6)
C23	1989(3)	-2209(2)	5627.3(17)	35.4(7)
C24	958(3)	-2515(2)	5700(2)	42.2(9)
C25	254(3)	-2558(2)	5091(2)	42.2(9)
C26	135(3)	-1802(2)	4787(2)	38.7(8)
C27	1158(3)	-1484(2)	4706.2(16)	30.0(7)
C28	1078(3)	-721(2)	4390.9(17)	35.4(7)
C29	317(3)	-200(2)	4593(2)	46.5(10)
C30	421(5)	585(3)	4365(3)	61.6(13)
C31	853(4)	-837(3)	3706.0(19)	43.8(9)
C32	1511(4)	-728(3)	2769(2)	51.4(10)
C33	2325(8)	-296(5)	2535(3)	85(2)
C34	1646(4)	-1547(3)	2646(3)	58.3(13)
C35	1167(6)	-2491(5)	1915(4)	83(2)
C36	117(5)	-2751(4)	1852(3)	72.3(16)
C37	-380(7)	-3039(6)	1286(4)	90(2)
C38	-1342(7)	-3243(6)	1196(4)	84(2)
C39	-1916(6)	-3156(4)	1647(4)	76.2(17)
C40	-1457(6)	-2877(4)	2196(3)	70.5(15)
C41	-462(6)	-2676(5)	2303(3)	74.0(17)
O1A	4871(3)	866(2)	-1905.9(14)	53.2(9)
O2A	4405(3)	1258.9(19)	-1034.6(13)	44.8(7)
O3A	5155(3)	-441(2)	-337.9(16)	54.4(8)
O4A	5333(3)	2222(2)	1095.7(14)	46.3(7)
O5A	2748(3)	2437.6(19)	410.3(14)	46.2(7)
O6A	3982(3)	3145(2)	2912.7(13)	48.2(7)
O7A	5824(3)	4012(3)	2290(2)	70.3(12)

O8A	6781(3)	3874(2)	3196.4(14)	51.4(8)
N1A	4014(4)	127(2)	-1414.0(16)	56.1(11)
N2A	3931(3)	-82(2)	167.5(16)	42.8(8)
N3A	4642(3)	2072(2)	110.9(15)	40.2(7)
N4A	3388(3)	3584(2)	641.6(14)	39.1(7)
N5A	5007(3)	2665(2)	2323.8(15)	43.1(8)
C1A	6147(5)	1811(4)	-1555(3)	64.3(14)
C2A	5676(6)	1387(4)	-2642(3)	70.4(17)
C3A	4460(5)	2144(3)	-2180(3)	58.2(12)
C4A	5297(4)	1573(3)	-2057(2)	51.3(11)
C5A	4435(4)	788(3)	-1412.8(18)	45.3(9)
C6A	3520(5)	-109(3)	-922.5(19)	53.8(12)
C7A	2997(7)	-850(4)	-1084(3)	81(2)
C8A	4294(4)	-223(2)	-335.0(19)	43.1(9)
C9A	4547(4)	-176(3)	778.0(18)	44.6(9)
C10A	3960(5)	-624(3)	1166(2)	56.4(12)
C11A	4540(7)	-700(4)	1815(2)	69.7(17)
C12A	4753(6)	70(4)	2093(2)	68.0(16)
C13A	5382(5)	510(3)	1720(2)	60.0(13)
C14A	4839(4)	586(3)	1058.0(17)	42.6(9)
C15A	5462(3)	1025(3)	654.3(18)	41.1(8)
C16A	6611(4)	942(4)	815(3)	59.3(12)
C17A	7129(5)	1250(5)	310(4)	76.6(18)
C18A	5147(3)	1833(2)	647.3(18)	38.6(8)
C19A	4266(4)	2817(2)	42.2(17)	43.5(9)
C20A	3911(5)	3000(3)	-625(2)	56.1(12)
C21A	3393(3)	2918(2)	390.9(16)	39.1(8)
C22A	2634(3)	3840(2)	987.8(17)	38.4(8)
C23A	2401(4)	4644(3)	804(2)	52.4(11)
C24A	1692(5)	5008(3)	1181(3)	56.6(12)
C25A	2180(5)	4965(3)	1848(3)	59.2(13)
C26A	2352(4)	4156(3)	2042(2)	50.5(10)
C27A	3056(3)	3777(2)	1669.8(17)	38.6(8)
C28A	3282(3)	2955(2)	1852.2(16)	38.0(8)
C29A	2357(3)	2509(3)	1946.7(19)	44.5(9)
C30A	2583(5)	1681(3)	1965(2)	54.5(11)
C31A	4107(3)	2931(2)	2418.4(18)	39.6(8)
C32A	5909(3)	2823(3)	2756.4(17)	41.0(8)
C33A	6792(4)	2364(3)	2618(2)	52.8(11)
C34A	6149(3)	3642(3)	2713.1(18)	42.1(9)
C35A	7171(5)	4619(3)	3157(2)	53.2(11)
C36A	8063(4)	4594(3)	2863(2)	55.6(12)
C37A	8749(6)	3999(4)	2966(4)	74.3(17)
C38A	9600(6)	3985(5)	2724(5)	88(2)
C39A	9767(7)	4572(5)	2361(5)	90(2)
C40A	9158(6)	5159(4)	2269(5)	81(2)
C41A	8291(5)	5155(4)	2523(3)	63.1(13)
O9	3127(5)	1406(3)	5857(2)	36.9(12)
C42	2184(6)	1526(4)	6037(3)	56(2)
C43	2124(6)	2295(4)	6236(3)	54.8(19)
O9A	3444(11)	1358(6)	5938(5)	30(3)

C42A	2774(11)	1547(10)	6326(7)	44(5)
C43A	1711(13)	1477(17)	6029(12)	74(7)
O10	2359(3)	993(2)	193(2)	63.7(10)
C44A	1345(18)	1010(20)	310(20)	98(2)
C45A	1220(20)	384(15)	701(14)	98(2)
C44	1369(10)	940(10)	340(11)	98(2)
C45	816(11)	352(9)	-34(7)	98(2)
C46	7270(30)	-836(18)	3827(12)	148(2)
C47	6930(20)	-970(20)	3166(12)	148(2)
C48	7600(20)	-1030(20)	2778(10)	148(2)
C49	7490(20)	-1020(20)	2088(10)	148(2)
C50	8390(30)	-760(30)	1831(13)	148(2)
C51	8360(20)	-960(20)	1198(13)	148(2)
C52	9340(20)	-580(20)	1045(16)	148(2)
C46A	7480(30)	-560(30)	3670(18)	148(2)
C47A	7730(30)	-260(20)	3094(16)	148(2)
C48A	7780(30)	-700(20)	2655(17)	148(2)
C49A	8360(30)	-340(20)	2197(18)	148(2)
C50A	8410(40)	-820(30)	1677(19)	148(2)
C51A	8860(30)	-590(30)	1180(18)	148(2)
C52A	9020(30)	-1110(20)	659(17)	148(2)
C53	9380(30)	1160(20)	2711(13)	148(2)
C54	9560(30)	1291(14)	2086(13)	148(2)
C55	9490(30)	1976(14)	1826(12)	148(2)
C56	9440(30)	2214(16)	1168(11)	148(2)
C57	9530(30)	3022(15)	1040(11)	148(2)
C58	9680(20)	3185(19)	424(13)	148(2)
C59	8580(20)	3130(20)	45(13)	148(2)
C53A	9270(30)	1530(20)	2571(14)	148(2)
C54A	9380(30)	2173(17)	2181(11)	148(2)
C55A	9360(30)	2102(15)	1572(10)	148(2)
C56A	9650(20)	2630(20)	1100(12)	148(2)
C57A	8830(20)	2840(20)	578(11)	148(2)
C58A	9110(30)	2760(20)	-15(12)	148(2)
C59A	8390(30)	3306(14)	-423(11)	148(2)

Table S51. Anisotropic Displacement Parameters ($Å^2 \times 10^3$) for Gellman153. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	$\mathbf{U_{11}}$	$\mathbf{U_{22}}$	$\mathbf{U_{33}}$	$\mathbf{U_{23}}$	$\mathbf{U_{13}}$	$\mathbf{U_{12}}$
O1	37.7(15)	44.1(18)	59.4(18)	8.6(14)	-1.6(13)	-8.3(12)
O2	41.2(15)	32.4(15)	60.4(18)	-0.3(13)	-1.1(12)	-8.6(11)
O3	43.1(14)	31.9(14)	42.9(13)	0.0(11)	10.7(11)	-8.2(11)
O4	41.0(13)	28.3(13)	35.4(12)	-4.1(10)	4.1(10)	-4.4(10)
O5	37.6(12)	21.4(12)	41.2(12)	-0.9(10)	11.9(10)	-2.5(9)
O6	54(2)	77(3)	56.7(19)	9.3(19)	-15.5(16)	-23.1(19)
O7	123(5)	63(3)	91(4)	9(3)	-17(3)	14(3)
O8	76(3)	82(3)	65(2)	-13(2)	4(2)	-15(2)
N1	46.0(19)	37(2)	52.0(19)	0.2(15)	-3.3(15)	-10.4(14)

N2	37.2(15)	32.1(16)	33.5(14)	-0.3(12)	11.3(11)	-5.4(12)
N3	30.7(13)	26.8(15)	32.4(13)	0.5(11)	6.6(10)	-3.9(10)
N4	26.8(13)	22.9(14)	34.8(13)	0.5(10)	5.8(10)	-1.2(10)
N5	51(2)	54(2)	36.6(17)	0.0(15)	-1.6(14)	-6.4(17)
C1	72(4)	71(4)	64(3)	1(3)	20(3)	20(3)
C2	43(3)	74(4)	89(4)	36(3)	7(3)	2(2)
C3	55(3)	57(3)	65(3)	25(2)	0(2)	-4(2)
C4	44(2)	52(3)	56(2)	19(2)	5.1(18)	2.4(19)
C5	38.3(19)	36(2)	42.7(18)	4.1(15)	-1.3(15)	-8.0(15)
C6	46(2)	30.9(19)	37.8(17)	1.2(14)	6.4(15)	-6.6(15)
C7	79(3)	40(2)	40(2)	-7.7(17)	7(2)	-1(2)
C8	37.9(17)	22.2(16)	37.6(16)	0.4(12)	7.5(13)	-4.6(12)
C9	35.3(17)	27.6(18)	35.9(16)	4.3(13)	9.2(13)	-2.9(13)
C10	42(2)	33(2)	50(2)	2.0(16)	11.3(16)	1.2(15)
C11	42(2)	43(2)	52(2)	10.1(18)	6.6(17)	7.6(16)
C12	37.3(19)	47(2)	44.4(19)	8.2(17)	5.9(15)	3.1(16)
C13	38.2(18)	39(2)	34.6(16)	0.0(14)	4.0(14)	0.0(15)
C14	33.5(16)	31.5(18)	30.6(15)	4.0(13)	10.9(12)	0.3(13)
C15	32.8(16)	30.1(18)	31.5(15)	3.1(12)	9.8(12)	1.2(13)
C16	45(2)	45(2)	37.2(18)	7.0(16)	19.4(15)	4.4(16)
C17	54(3)	69(3)	56(3)	11(2)	30(2)	14(2)
C18	32.7(16)	26.3(17)	29.8(15)	-1.1(12)	9.7(12)	0.2(12)
C19	28.9(16)	27.5(18)	37.1(16)	1.5(13)	4.6(13)	-5.5(12)
C20	34.1(18)	42(2)	45(2)	12.2(16)	-1.5(15)	-8.7(15)
C21	30.2(15)	20.5(16)	30.8(14)	1.0(11)	6.0(12)	-3.1(11)
C22	29.2(15)	24.9(17)	32.8(15)	-0.8(12)	5.0(12)	-0.6(12)
C23	40.9(18)	24.1(17)	40.0(17)	4.0(13)	4.1(14)	-5.5(13)
C24	50(2)	29.7(19)	47(2)	2.9(15)	9.3(17)	-13.4(16)
C25	39.9(19)	32(2)	55(2)	-4.5(17)	8.7(16)	-12.0(15)
C26	29.6(17)	30.9(19)	55(2)	-1.9(16)	6.0(14)	-5.0(13)
C27	27.1(15)	26.4(17)	36.3(16)	-1.8(12)	5.4(12)	-1.4(12)
C28	31.3(16)	31.8(19)	40.4(18)	2.6(14)	-0.5(13)	-2.2(13)
C29	37.5(19)	31(2)	72(3)	8.5(19)	11.5(18)	5.9(15)
C30	65(3)	31(2)	88(4)	15(2)	12(3)	9(2)
C31	47(2)	38(2)	40.4(19)	3.0(15)	-8.1(16)	-1.9(16)
C32	64(3)	45(3)	41(2)	3.4(18)	-0.5(18)	-4(2)
C33	132(7)	75(5)	51(3)	-4(3)	27(3)	-37(5)
C34	53(3)	58(3)	58(3)	7(2)	-6(2)	-11(2)
C35	76(4)	80(5)	94(5)	-25(4)	17(3)	1(3)
C36	70(4)	64(4)	79(4)	-21(3)	6(3)	4(3)
C37	97(5)	100(6)	71(4)	-30(4)	13(4)	-13(5)
C38	84(5)	85(5)	81(4)	-12(4)	11(4)	-4(4)
C39	80(4)	60(4)	85(4)	10(3)	5(3)	-8(3)
C40	77(4)	62(4)	75(4)	2(3)	20(3)	-2(3)
C41	72(4)	72(4)	77(4)	-17(3)	12(3)	-7(3)
O1A	93(3)	37.6(17)	34.8(14)	-1.3(12)	27.2(16)	5.6(16)
O2A	66.3(19)	37.3(16)	33.5(13)	-7.8(11)	16.4(12)	-3.3(13)
O3A	79(2)	40.0(18)	49.2(17)	-4.5(13)	26.2(16)	8.3(15)
O4A	59.8(18)	43.2(17)	36.1(14)	-9.2(12)	9.5(12)	-2.9(13)
O5A	54.5(17)	37.8(16)	45.5(14)	-5.3(12)	7.4(12)	-2.1(13)
O6A	59.0(18)	56(2)	31.0(13)	2.5(12)	11.8(12)	5.2(14)

O7A	68(2)	67(3)	69(2)	38(2)	-6.0(18)	-11.3(19)
O8A	80(2)	36.7(17)	35.6(13)	1.5(12)	6.0(14)	2.0(15)
N1A	103(3)	39(2)	29.4(15)	-7.5(14)	20.6(18)	-6(2)
N2A	54.0(19)	43(2)	32.3(15)	-1.4(13)	9.8(14)	-4.5(15)
N3A	57(2)	33.4(18)	31.7(14)	-5.7(12)	10.6(13)	-1.3(14)
N4A	52.5(19)	32.9(17)	32.1(14)	1.2(12)	8.1(13)	0.0(14)
N5A	52(2)	48(2)	28.3(13)	-2.8(13)	5.0(13)	8.4(15)
C1A	60(3)	77(4)	58(3)	11(3)	16(2)	-5(3)
C2A	98(4)	74(4)	47(3)	18(3)	34(3)	22(3)
C3A	80(3)	45(3)	53(2)	7(2)	19(2)	7(2)
C4A	69(3)	51(3)	38(2)	9.5(18)	20.4(19)	6(2)
C5A	68(3)	43(2)	26.6(16)	-1.8(15)	10.8(17)	-1.8(19)
C6A	79(3)	49(3)	32.5(19)	-3.1(17)	8.9(19)	-21(2)
C7A	128(6)	62(4)	47(3)	2(2)	-3(3)	-49(4)
C8A	65(3)	28.9(19)	37.5(19)	-5.3(14)	14.9(18)	-5.7(17)
C9A	66(3)	38(2)	30.1(17)	-0.8(15)	10.4(17)	4.8(18)
C10A	89(4)	41(2)	43(2)	2.6(18)	22(2)	-5(2)
C11A	120(5)	50(3)	42(2)	10(2)	24(3)	9(3)
C12A	117(5)	57(3)	29.2(19)	4(2)	12(2)	-6(3)
C13A	88(4)	53(3)	34(2)	-0.2(19)	-2(2)	0(3)
C14A	59(2)	38(2)	30.8(17)	-2.4(15)	7.6(16)	1.5(17)
C15A	50(2)	39(2)	33.7(16)	-4.6(15)	5.8(15)	2.8(16)
C16A	52(3)	54(3)	69(3)	-6(2)	5(2)	7(2)
C17A	56(3)	83(5)	94(5)	-13(4)	25(3)	-2(3)
C18A	47(2)	35(2)	35.3(17)	-6.8(15)	9.7(15)	-2.2(15)
C19A	70(3)	32(2)	30.3(17)	-0.6(14)	15.8(17)	-5.0(18)
C20A	98(4)	40(2)	32.1(18)	1.6(16)	17(2)	2(2)
C21A	58(2)	33(2)	25.9(14)	3.4(13)	5.9(14)	0.3(16)
C22A	45(2)	35(2)	34.8(17)	0.9(14)	6.7(14)	4.9(15)
C23A	67(3)	44(3)	44(2)	6.8(18)	3.7(19)	19(2)
C24A	66(3)	43(3)	59(3)	0(2)	8(2)	14(2)
C25A	79(3)	43(3)	58(3)	-6(2)	18(2)	14(2)
C26A	63(3)	45(3)	46(2)	1.1(18)	18.7(19)	15(2)
C27A	46(2)	38(2)	32.6(16)	-1.2(14)	9.0(14)	5.3(15)
C28A	43.7(19)	40(2)	30.8(15)	2.8(14)	8.2(14)	6.5(15)
C29A	49(2)	44(2)	40.2(19)	7.8(16)	9.1(16)	1.7(18)
C30A	70(3)	47(3)	46(2)	4.4(19)	12(2)	-5(2)
C31A	51(2)	35(2)	33.6(17)	2.9(14)	8.3(15)	1.7(16)
C32A	48(2)	43(2)	30.1(16)	2.3(15)	2.8(14)	6.1(17)
C33A	53(2)	57(3)	45(2)	-6(2)	-1.4(18)	12(2)
C34A	49(2)	48(2)	31.5(17)	8.7(16)	13.2(15)	10.4(17)
C35A	80(3)	39(2)	43(2)	-2.2(17)	17(2)	4(2)
C36A	58(3)	61(3)	45(2)	-9(2)	1.5(19)	-5(2)
C37A	83(4)	57(4)	77(4)	-1(3)	0(3)	7(3)
C38A	61(4)	68(4)	127(7)	-4(4)	-2(4)	17(3)
C39A	83(5)	68(5)	128(7)	-3(4)	41(5)	-9(4)
C40A	75(4)	61(4)	112(6)	10(4)	30(4)	-3(3)
C41A	66(3)	53(3)	70(3)	1(2)	10(3)	8(2)
O10	58(2)	43(2)	89(3)	-8.3(19)	10.9(19)	-6.7(16)

Table S52. Bond Lengths for Gellman153.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
O1	C4	1.465(7)	N4A	C21A	1.335(6)
O1	C5	1.355(6)	N4A	C22A	1.460(5)
O2	C5	1.224(5)	N5A	C31A	1.355(6)
O3	C8	1.235(5)	N5A	C32A	1.436(6)
O4	C18	1.233(5)	C1A	C4A	1.514(8)
O5	C21	1.244(4)	C2A	C4A	1.530(6)
O6	C31	1.229(6)	C3A	C4A	1.519(8)
O7	C34	1.198(8)	C6A	C7A	1.531(7)
O8	C34	1.304(7)	C6A	C8A	1.537(7)
O8	C35	1.462(10)	C9A	C10A	1.515(7)
N1	C5	1.339(7)	C9A	C14A	1.542(6)
N1	C6	1.452(6)	C10A	C11A	1.530(8)
N2	C8	1.339(5)	C11A	C12A	1.539(9)
N2	C9	1.468(5)	C12A	C13A	1.520(9)
N3	C18	1.344(5)	C13A	C14A	1.539(6)
N3	C19	1.449(5)	C14A	C15A	1.561(6)
N4	C21	1.331(5)	C15A	C16A	1.527(7)
N4	C22	1.461(4)	C15A	C18A	1.528(6)
N5	C31	1.335(7)	C16A	C17A	1.541(10)
N5	C32	1.455(6)	C19A	C20A	1.524(6)
C1	C4	1.514(9)	C19A	C21A	1.536(6)
C2	C4	1.530(7)	C22A	C23A	1.536(6)
C3	C4	1.534(7)	C22A	C27A	1.537(5)
C6	C7	1.534(6)	C23A	C24A	1.536(7)
C6	C8	1.528(5)	C24A	C25A	1.523(8)
C9	C10	1.525(5)	C25A	C26A	1.540(8)
C9	C14	1.538(5)	C26A	C27A	1.536(6)
C10	C11	1.531(6)	C27A	C28A	1.564(6)
C11	C12	1.514(7)	C28A	C29A	1.530(6)
C12	C13	1.529(6)	C28A	C31A	1.527(5)
C13	C14	1.541(5)	C29A	C30A	1.534(7)
C14	C15	1.554(5)	C32A	C33A	1.528(6)
C15	C16	1.542(5)	C32A	C34A	1.531(7)
C15	C18	1.521(5)	C35A	C36A	1.472(8)
C16	C17	1.521(6)	C36A	C37A	1.413(10)
C19	C20	1.532(5)	C36A	C41A	1.343(9)
C19	C21	1.534(5)	C37A	C38A	1.352(13)
C22	C23	1.531(5)	C38A	C39A	1.386(14)
C22	C27	1.544(5)	C39A	C40A	1.338(13)
C23	C24	1.529(5)	C40A	C41A	1.387(11)
C24	C25	1.516(6)	O9	C42	1.415(9)
C25	C26	1.531(6)	C42	C43	1.474(10)
C26	C27	1.533(5)	O9A	C42A	1.405(12)
C27	C28	1.551(5)	C42A	C43A	1.467(15)
C28	C29	1.523(6)	O10	C44A	1.432(12)
C28	C31	1.527(6)	O10	C44	1.432(11)
C29	C30	1.531(7)	C44A	C45A	1.469(16)
C32	C33	1.517(9)	C44	C45	1.474(16)
C32	C34	1.530(9)	C46	C47	1.49(3)

C35	C36	1.470(11)	C47	C48	1.367(17)
C36	C37	1.422(10)	C48	C49	1.53(2)
C36	C41	1.393(11)	C49	C50	1.51(3)
C37	C38	1.324(13)	C50	C51	1.46(3)
C38	C39	1.391(13)	C51	C52	1.57(3)
C39	C40	1.371(11)	C46A	C47A	1.50(5)
C40	C41	1.366(11)	C47A	C48A	1.28(3)
O1A	C4A	1.472(6)	C48A	C49A	1.54(5)
O1A	C5A	1.352(5)	C49A	C50A	1.47(6)
O2A	C5A	1.211(6)	C50A	C51A	1.42(6)
O3A	C8A	1.225(7)	C51A	C52A	1.55(6)
O4A	C18A	1.218(5)	C53	C54	1.49(3)
O5A	C21A	1.237(6)	C54	C55	1.371(17)
O6A	C31A	1.217(5)	C55	C56	1.53(2)
O7A	C34A	1.181(6)	C56	C57	1.50(3)
O8A	C34A	1.319(6)	C57	C58	1.46(3)
O8A	C35A	1.461(7)	C58	C59	1.57(3)
N1A	C5A	1.329(7)	C53A	C54A	1.49(3)
N1A	C6A	1.453(6)	C54A	C55A	1.369(17)
N2A	C8A	1.333(6)	C55A	C56A	1.53(2)
N2A	C9A	1.476(5)	C56A	C57A	1.50(3)
N3A	C18A	1.343(6)	C57A	C58A	1.46(3)
N3A	C19A	1.443(6)	C58A	C59A	1.57(3)

Table S53. Bond Angles for Gellman153.

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C5	O1	C4	120.5(4)	C3A	C4A	C2A	110.3(4)
C34	O8	C35	118.7(6)	O2A	C5A	O1A	125.2(5)
C5	N1	C6	120.2(4)	O2A	C5A	N1A	124.9(4)
C8	N2	C9	123.6(3)	N1A	C5A	O1A	109.9(4)
C18	N3	C19	121.2(3)	N1A	C6A	C7A	109.4(4)
C21	N4	C22	125.5(3)	N1A	C6A	C8A	111.1(5)
C31	N5	C32	121.6(4)	C7A	C6A	C8A	107.1(5)
O1	C4	C1	111.2(4)	O3A	C8A	N2A	123.9(4)
O1	C4	C2	101.7(5)	O3A	C8A	C6A	122.1(4)
O1	C4	C3	109.0(5)	N2A	C8A	C6A	114.0(4)
C1	C4	C2	112.1(5)	N2A	C9A	C10A	109.4(4)
C1	C4	C3	112.3(5)	N2A	C9A	C14A	109.5(4)
C2	C4	C3	110.1(4)	C10A	C9A	C14A	111.6(4)
O2	C5	O1	124.9(4)	C9A	C10A	C11A	111.7(5)
O2	C5	N1	124.1(4)	C10A	C11A	C12A	109.2(5)
N1	C5	O1	111.1(4)	C13A	C12A	C11A	109.6(5)
N1	C6	C7	109.8(4)	C12A	C13A	C14A	111.5(5)
N1	C6	C8	111.0(3)	C9A	C14A	C15A	110.2(3)
C8	C6	C7	109.5(3)	C13A	C14A	C9A	110.7(4)
O3	C8	N2	123.4(3)	C13A	C14A	C15A	113.4(4)
O3	C8	C6	121.7(3)	C16A	C15A	C14A	116.1(4)
N2	C8	C6	114.8(3)	C16A	C15A	C18A	111.3(4)

N2	C9	C10	108.9(3)	C18A	C15A	C14A	108.7(3)
N2	C9	C14	109.4(3)	C15A	C16A	C17A	111.3(5)
C10	C9	C14	112.3(3)	O4A	C18A	N3A	123.2(4)
C9	C10	C11	112.2(3)	O4A	C18A	C15A	122.3(4)
C12	C11	C10	110.1(4)	N3A	C18A	C15A	114.5(3)
C11	C12	C13	111.0(4)	N3A	C19A	C20A	110.6(4)
C12	C13	C14	112.5(3)	N3A	C19A	C21A	110.1(3)
C9	C14	C13	110.3(3)	C20A	C19A	C21A	110.1(4)
C9	C14	C15	109.9(3)	O5A	C21A	N4A	124.6(4)
C13	C14	C15	114.2(3)	O5A	C21A	C19A	122.6(4)
C16	C15	C14	115.0(3)	N4A	C21A	C19A	112.8(4)
C18	C15	C14	109.3(3)	N4A	C22A	C23A	106.7(4)
C18	C15	C16	110.9(3)	N4A	C22A	C27A	110.0(3)
C17	C16	C15	110.9(3)	C23A	C22A	C27A	111.0(4)
O4	C18	N3	122.0(3)	C24A	C23A	C22A	112.3(4)
O4	C18	C15	122.8(3)	C25A	C24A	C23A	108.7(4)
N3	C18	C15	115.2(3)	C24A	C25A	C26A	110.0(5)
N3	C19	C20	110.1(3)	C27A	C26A	C25A	110.4(4)
N3	C19	C21	109.7(3)	C22A	C27A	C28A	110.5(3)
C20	C19	C21	109.2(3)	C26A	C27A	C22A	111.0(4)
O5	C21	N4	124.0(3)	C26A	C27A	C28A	113.3(4)
O5	C21	C19	121.9(3)	C29A	C28A	C27A	114.7(3)
N4	C21	C19	114.1(3)	C31A	C28A	C27A	108.8(3)
N4	C22	C23	108.2(3)	C31A	C28A	C29A	110.7(3)
N4	C22	C27	110.7(3)	C28A	C29A	C30A	111.2(4)
C23	C22	C27	111.0(3)	O6A	C31A	N5A	122.2(4)
C24	C23	C22	111.6(3)	O6A	C31A	C28A	123.4(4)
C25	C24	C23	110.6(3)	N5A	C31A	C28A	114.4(3)
C24	C25	C26	110.7(3)	N5A	C32A	C33A	110.1(4)
C25	C26	C27	111.6(3)	N5A	C32A	C34A	108.2(4)
C22	C27	C28	110.9(3)	C33A	C32A	C34A	109.7(4)
C26	C27	C22	110.2(3)	O7A	C34A	O8A	124.7(5)
C26	C27	C28	113.7(3)	O7A	C34A	C32A	123.6(5)
C29	C28	C27	114.5(3)	O8A	C34A	C32A	111.7(3)
C29	C28	C31	111.4(4)	O8A	C35A	C36A	109.2(4)
C31	C28	C27	108.7(3)	C37A	C36A	C35A	120.7(6)
C28	C29	C30	111.9(4)	C41A	C36A	C35A	121.5(6)
O6	C31	N5	121.7(4)	C41A	C36A	C37A	117.6(6)
O6	C31	C28	123.4(5)	C38A	C37A	C36A	121.6(8)
N5	C31	C28	114.9(4)	C37A	C38A	C39A	117.5(7)
N5	C32	C33	110.6(4)	C40A	C39A	C38A	123.0(8)
N5	C32	C34	109.6(4)	C39A	C40A	C41A	117.9(7)
C33	C32	C34	108.9(6)	C36A	C41A	C40A	122.3(6)
O7	C34	O8	124.9(7)	O9	C42	C43	109.5(6)
O7	C34	C32	123.9(6)	O9A	C42A	C43A	112.2(13)
O8	C34	C32	110.9(5)	O10	C44A	C45A	107.9(16)
O8	C35	C36	109.9(6)	O10	C44	C45	108.0(12)
C37	C36	C35	119.4(7)	C48	C47	C46	121.9(19)
C41	C36	C35	123.5(6)	C47	C48	C49	134.0(19)
C41	C36	C37	116.9(7)	C50	C49	C48	117.3(18)
C38	C37	C36	121.8(8)	C51	C50	C49	115(2)

C37	C38	C39	120.7(8)	C50	C51	C52	103.6(17)
C40	C39	C38	118.7(7)	C48A	C47A	C46A	119(3)
C41	C40	C39	121.5(7)	C47A	C48A	C49A	110(3)
C40	C41	C36	120.3(7)	C50A	C49A	C48A	112(4)
C5A	O1A	C4A	121.7(4)	C51A	C50A	C49A	123(5)
C34A	O8A	C35A	115.5(4)	C50A	C51A	C52A	123(4)
C5A	N1A	C6A	121.2(4)	C55	C54	C53	122.3(19)
C8A	N2A	C9A	122.3(4)	C54	C55	C56	130.9(18)
C18A	N3A	C19A	120.7(3)	C57	C56	C55	118.0(18)
C21A	N4A	C22A	124.8(4)	C58	C57	C56	114(2)
C31A	N5A	C32A	119.3(3)	C57	C58	C59	103.0(17)
O1A	C4A	C1A	110.5(4)	C55A	C54A	C53A	121.6(19)
O1A	C4A	C2A	102.1(5)	C54A	C55A	C56A	132.1(18)
O1A	C4A	C3A	109.4(4)	C57A	C56A	C55A	117.4(17)
C1A	C4A	C2A	112.3(5)	C58A	C57A	C56A	114(2)
C1A	C4A	C3A	111.9(5)	C57A	C58A	C59A	103.3(17)

Table S54. Hydrogen Bonds for Gellman153.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
N1	H1	O7[1]	0.88	2.09	2.893(7)	151.7
N2	H2	O9	0.88	2.16	2.945(5)	148.0
N3	H3	O2	0.88	2.05	2.913(4)	165.8
N4	H4	O3[2]	0.88	1.92	2.784(4)	165.7
N5	H5	O4	0.88	2.08	2.951(5)	173.0
N1A	H1AA	O7A[3]	0.88	1.99	2.860(5)	169.6
N2A	H2AA	O10	0.88	2.12	2.886(6)	145.2
N3A	H3AA	O2A	0.88	2.07	2.934(4)	167.5
N4A	H4A	O3A[4]	0.88	1.95	2.816(5)	169.7
N5A	H5A	O4A	0.88	2.16	2.982(5)	154.3
O9	H9B	O5	0.84	1.92	2.725(5)	160.1
O10	H10	O5A	0.84	1.88	2.704(5)	166.8

[1]1-X,1/2+Y,1-Z; [2]1-X,-1/2+Y,1-Z; [3]1-X,-1/2+Y,-Z; [4]1-X,1/2+Y,-Z

Table S55. Torsion Angles for Gellman153.

A	B	C	D	Angle/°	A	B	C	D	Angle/°
O8	C35	C36	C37	121.3(9)	N4A	C22A	C23A	C24A	-174.4(4)
O8	C35	C36	C41	-53.1(11)	N4A	C22A	C27A	C26A	170.9(4)
N1	C6	C8	O3	-35.1(5)	N4A	C22A	C27A	C28A	-62.6(4)
N1	C6	C8	N2	148.2(3)	N5A	C32A	C34A	O7A	-19.4(6)
N2	C9	C10	C11	-175.6(3)	N5A	C32A	C34A	O8A	162.4(4)
N2	C9	C14	C13	172.7(3)	C4A	O1A	C5A	O2A	-4.9(8)
N2	C9	C14	C15	-60.6(3)	C4A	O1A	C5A	N1A	173.1(5)
N3	C19	C21	O5	-35.1(4)	C5A	O1A	C4A	C1A	61.3(6)
N3	C19	C21	N4	146.8(3)	C5A	O1A	C4A	C2A	-179.1(5)
N4	C22	C23	C24	-177.3(3)	C5A	O1A	C4A	C3A	-62.3(6)

N4	C22	C27	C26	174.8(3)	C5A	N1A	C6A	C7A	173.0(6)
N4	C22	C27	C28	-58.5(4)	C5A	N1A	C6A	C8A	-68.9(7)
N5	C32	C34	O7	-17.9(9)	C6A	N1A	C5A	O1A	178.0(5)
N5	C32	C34	O8	156.1(5)	C6A	N1A	C5A	O2A	-4.0(9)
C4	O1	C5	O2	-12.6(7)	C7A	C6A	C8A	O3A	86.5(6)
C4	O1	C5	N1	167.8(4)	C7A	C6A	C8A	N2A	-91.8(6)
C5	O1	C4	C1	64.5(6)	C8A	N2A	C9A	C10A	-130.3(5)
C5	O1	C4	C2	-176.1(4)	C8A	N2A	C9A	C14A	107.1(5)
C5	O1	C4	C3	-59.8(6)	C9A	N2A	C8A	O3A	0.3(7)
C5	N1	C6	C7	171.7(4)	C9A	N2A	C8A	C6A	178.5(4)
C5	N1	C6	C8	-67.1(5)	C9A	C10A	C11A	C12A	58.7(7)
C6	N1	C5	O1	178.1(4)	C9A	C14A	C15A	C16A	-91.8(5)
C6	N1	C5	O2	-1.5(7)	C9A	C14A	C15A	C18A	141.9(4)
C7	C6	C8	O3	86.3(5)	C10A	C9A	C14A	C13A	52.5(6)
C7	C6	C8	N2	-90.4(4)	C10A	C9A	C14A	C15A	178.8(4)
C8	N2	C9	C10	-127.4(4)	C10A	C11A	C12A	C13A	-59.9(8)
C8	N2	C9	C14	109.5(4)	C11A	C12A	C13A	C14A	58.9(7)
C9	N2	C8	O3	0.8(6)	C12A	C13A	C14A	C9A	-54.7(6)
C9	N2	C8	C6	177.5(3)	C12A	C13A	C14A	C15A	-179.2(5)
C9	C10	C11	C12	56.4(5)	C13A	C14A	C15A	C16A	32.9(6)
C9	C14	C15	C16	-93.3(4)	C13A	C14A	C15A	C18A	-93.3(5)
C9	C14	C15	C18	141.1(3)	C14A	C9A	C10A	C11A	-55.4(6)
C10	C9	C14	C13	51.6(4)	C14A	C15A	C16A	C17A	167.2(5)
C10	C9	C14	C15	178.4(3)	C14A	C15A	C18A	O4A	67.7(5)
C10	C11	C12	C13	-57.1(5)	C14A	C15A	C18A	N3A	-111.6(4)
C11	C12	C13	C14	56.8(5)	C16A	C15A	C18A	O4A	-61.3(6)
C12	C13	C14	C9	-53.1(4)	C16A	C15A	C18A	N3A	119.4(4)
C12	C13	C14	C15	-177.4(3)	C18A	N3A	C19A	C20A	168.8(4)
C13	C14	C15	C16	31.3(5)	C18A	N3A	C19A	C21A	-69.3(5)
C13	C14	C15	C18	-94.3(3)	C18A	C15A	C16A	C17A	-67.8(6)
C14	C9	C10	C11	-54.3(4)	C19A	N3A	C18A	O4A	-1.4(7)
C14	C15	C16	C17	167.5(4)	C19A	N3A	C18A	C15A	177.8(4)
C14	C15	C18	O4	69.9(4)	C20A	C19A	C21A	O5A	83.4(5)
C14	C15	C18	N3	-107.5(3)	C20A	C19A	C21A	N4A	-93.8(4)
C16	C15	C18	O4	-57.9(5)	C21A	N4A	C22A	C23A	-139.9(4)
C16	C15	C18	N3	124.7(3)	C21A	N4A	C22A	C27A	99.6(4)
C18	N3	C19	C20	176.9(3)	C22A	N4A	C21A	O5A	1.4(6)
C18	N3	C19	C21	-62.8(4)	C22A	N4A	C21A	C19A	178.6(4)
C18	C15	C16	C17	-67.8(5)	C22A	C23A	C24A	C25A	58.0(6)
C19	N3	C18	O4	-1.6(5)	C22A	C27A	C28A	C29A	-81.2(4)
C19	N3	C18	C15	175.8(3)	C22A	C27A	C28A	C31A	154.2(3)
C20	C19	C21	O5	85.7(4)	C23A	C22A	C27A	C26A	53.0(5)
C20	C19	C21	N4	-92.5(4)	C23A	C22A	C27A	C28A	179.5(4)
C21	N4	C22	C23	-140.3(3)	C23A	C24A	C25A	C26A	-60.3(7)
C21	N4	C22	C27	97.8(4)	C24A	C25A	C26A	C27A	60.4(6)
C22	N4	C21	O5	-2.2(5)	C25A	C26A	C27A	C22A	-56.2(6)
C22	N4	C21	C19	175.9(3)	C25A	C26A	C27A	C28A	178.8(4)
C22	C23	C24	C25	56.9(5)	C26A	C27A	C28A	C29A	44.0(5)
C22	C27	C28	C29	-84.3(4)	C26A	C27A	C28A	C31A	-80.6(5)
C22	C27	C28	C31	150.4(3)	C27A	C22A	C23A	C24A	-54.6(6)
C23	C22	C27	C26	54.6(4)	C27A	C28A	C29A	C30A	165.7(3)

C23	C22	C27	C28	-178.7(3)	C27A	C28A	C31A	O6A	67.9(5)
C23	C24	C25	C26	-57.1(5)	C27A	C28A	C31A	N5A	-110.5(4)
C24	C25	C26	C27	57.3(5)	C29A	C28A	C31A	O6A	-59.0(6)
C25	C26	C27	C22	-55.6(4)	C29A	C28A	C31A	N5A	122.5(4)
C25	C26	C27	C28	179.2(3)	C31A	N5A	C32A	C33A	170.2(4)
C26	C27	C28	C29	40.6(4)	C31A	N5A	C32A	C34A	-69.9(5)
C26	C27	C28	C31	-84.7(4)	C31A	C28A	C29A	C30A	-70.7(5)
C27	C22	C23	C24	-55.7(4)	C32A	N5A	C31A	O6A	-17.8(7)
C27	C28	C29	C30	168.6(4)	C32A	N5A	C31A	C28A	160.7(4)
C27	C28	C31	O6	64.3(6)	C33A	C32A	C34A	O7A	100.8(5)
C27	C28	C31	N5	-115.3(4)	C33A	C32A	C34A	O8A	-77.4(4)
C29	C28	C31	O6	-62.8(6)	C34A	O8A	C35A	C36A	-85.5(5)
C29	C28	C31	N5	117.6(4)	C35A	O8A	C34A	O7A	-6.8(7)
C31	N5	C32	C33	169.2(6)	C35A	O8A	C34A	C32A	171.3(4)
C31	N5	C32	C34	-70.6(6)	C35A	C36A	C37A	C38A	-176.5(7)
C31	C28	C29	C30	-67.5(5)	C35A	C36A	C41A	C40A	176.4(7)
C32	N5	C31	O6	-3.9(8)	C36A	C37A	C38A	C39A	-1.3(13)
C32	N5	C31	C28	175.7(4)	C37A	C36A	C41A	C40A	0.8(10)
C33	C32	C34	O7	103.3(8)	C37A	C38A	C39A	C40A	3.9(15)
C33	C32	C34	O8	-82.8(6)	C38A	C39A	C40A	C41A	-4.0(16)
C34	O8	C35	C36	107.2(8)	C39A	C40A	C41A	C36A	1.6(13)
C35	O8	C34	O7	1.4(10)	C41A	C36A	C37A	C38A	-0.9(10)
C35	O8	C34	C32	-172.4(6)	C44A	O10	C44	C45	106(32)
C35	C36	C37	C38	-176.2(10)	C44	O10	C44A	C45A	3(28)
C35	C36	C41	C40	174.8(8)	C46	C47	C48	C49	-166(4)
C36	C37	C38	C39	2.5(17)	C47	C48	C49	C50	153(5)
C37	C36	C41	C40	0.3(13)	C48	C49	C50	C51	163(4)
C37	C38	C39	C40	-2.1(14)	C49	C50	C51	C52	177(4)
C38	C39	C40	C41	0.9(12)	C46A	C47A	C48A	C49A	-162(3)
C39	C40	C41	C36	0.0(13)	C47A	C48A	C49A	C50A	-178(4)
C41	C36	C37	C38	-1.5(15)	C48A	C49A	C50A	C51A	177(5)
O8A	C35A	C36A	C37A	-35.9(7)	C49A	C50A	C51A	C52A	174(4)
O8A	C35A	C36A	C41A	148.7(5)	C53	C54	C55	C56	-166(4)
N1A	C6A	C8A	O3A	-32.9(6)	C54	C55	C56	C57	-170(4)
N1A	C6A	C8A	N2A	148.8(4)	C55	C56	C57	C58	169(3)
N2A	C9A	C10A	C11A	-176.7(4)	C56	C57	C58	C59	80(4)
N2A	C9A	C14A	C13A	173.8(4)	C53A	C54A	C55A	C56A	-166(3)
N2A	C9A	C14A	C15A	-59.9(5)	C54A	C55A	C56A	C57A	-120(4)
N3A	C19A	C21A	O5A	-38.7(5)	C55A	C56A	C57A	C58A	-130(3)
N3A	C19A	C21A	N4A	144.0(4)	C56A	C57A	C58A	C59A	-157(3)

Table S56. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for Gellman153.

Atom	***x***	***y***	***z***	**U(eq)**
H1	7064	2556	6219	56
H2	3782	2286	5408	40
H3	4938	575	5207	36
H4	3298	-1551	5133	34

H5	2114	-385	3621	58
H1A	7259	311	5248	102
H1B	8357	-60	5339	102
H1C	8189	754	5067	102
H2A	9601	1208	5911	104
H2B	9740	394	6185	104
H2C	9498	1062	6600	104
H3A	8029	455	6857	91
H3B	8142	-257	6456	91
H3C	7104	196	6347	91
H6	5121	2135	6135	46
H7A	5651	3197	6694	80
H7B	4768	3436	6150	80
H7C	5925	3598	6110	80
H9	3983	2709	4239	39
H10A	2848	3467	4627	49
H10B	2222	2772	4794	49
H11A	1333	3306	3896	55
H11B	2309	3241	3582	55
H12A	1193	2269	3243	52
H12B	1196	2018	3927	52
H13A	2911	1946	3331	45
H13B	2296	1248	3504	45
H14	2863	1459	4542	37
H15	4762	1445	4566	37
H16A	4150	961	3338	49
H16B	4624	1761	3501	49
H17A	5911	943	3339	86
H17B	5658	411	3860	86
H17C	6131	1208	4030	86
H19	4530	-847	4972	38
H20A	5760	-526	5822	62
H20B	4907	-1007	6051	62
H20C	4922	-133	6134	62
H22	1621	-1095	5591	35
H23A	2315	-2554	5381	42
H23B	2429	-2167	6031	42
H24A	1046	-3012	5883	51
H24B	657	-2194	5975	51
H25A	-417	-2740	5148	51
H25B	529	-2911	4827	51
H26A	-192	-1461	5036	46
H26B	-312	-1845	4386	46
H27	1456	-1832	4442	36
H28	1759	-483	4494	42
H29A	420	-197	5040	56
H29B	-377	-381	4437	56
H30A	400	577	3927	92
H30B	1067	794	4568	92
H30C	-137	887	4455	92
H32	831	-565	2552	62

H33A	2249	229	2615	127
H33B	2258	-374	2097	127
H33C	2994	-464	2739	127
H35A	1611	-2788	2225	100
H35B	1403	-2550	1525	100
H37	-12	-3086	965	107
H38	-1644	-3451	818	101
H39	-2612	-3287	1577	91
H40	-1841	-2823	2509	85
H41	-162	-2484	2688	89
H1AA	4035	-175	-1718	67
H2AA	3302	71	134	51
H3AA	4539	1772	-202	48
H4A	3874	3891	595	47
H5A	5040	2399	2001	52
H1AB	5873	1917	-1189	96
H1AC	6469	2254	-1681	96
H1AD	6649	1416	-1471	96
H2AB	6284	1080	-2547	106
H2AC	5839	1843	-2837	106
H2AD	5148	1120	-2917	106
H3AB	3908	1959	-2490	87
H3AC	4727	2600	-2322	87
H3AD	4204	2242	-1806	87
H6A	3008	266	-855	65
H7AA	3490	-1205	-1183	122
H7AB	2450	-787	-1435	122
H7AC	2716	-1030	-739	122
H9A	5180	-448	741	54
H10C	3825	-1119	986	68
H10D	3301	-384	1170	68
H11C	5186	-963	1816	84
H11D	4135	-989	2059	84
H12C	4106	328	2099	82
H12D	5123	26	2515	82
H13C	6042	262	1732	72
H13D	5516	1005	1900	72
H14A	4197	863	1060	51
H15A	5252	837	231	49
H16C	6785	415	878	71
H16D	6866	1206	1197	71
H17D	6848	1009	-75	115
H17E	7858	1156	411	115
H17F	7009	1782	272	115
H19A	4827	3160	214	52
H20D	4444	2873	-852	84
H20E	3764	3528	-668	84
H20F	3298	2719	-783	84
H22A	2005	3538	882	46
H23C	3043	4924	856	63
H23D	2083	4663	371	63

H24C	1575	5529	1059	68
H24D	1031	4752	1113	68
H25C	1737	5206	2097	71
H25D	2836	5229	1915	71
H26C	2656	4132	2478	61
H26D	1694	3895	1982	61
H27A	3716	4046	1750	46
H28A	3561	2716	1517	46
H29C	1784	2613	1613	53
H29D	2156	2660	2331	53
H30D	2774	1530	1583	82
H30E	1979	1409	2024	82
H30F	3141	1575	2301	82
H32A	5793	2707	3174	49
H33D	6639	1840	2650	79
H33E	7407	2486	2909	79
H33F	6898	2473	2207	79
H35C	7360	4831	3568	64
H35D	6642	4934	2920	64
H37A	8609	3599	3210	89
H38A	10066	3589	2801	106
H39A	10339	4556	2167	108
H40A	9316	5567	2038	97
H41A	7843	5563	2453	76
H9B	3119	997	5683	55
H42A	2114	1188	6374	67
H42B	1626	1423	5694	67
H43A	2743	2421	6521	82
H43B	1538	2353	6434	82
H43C	2052	2621	5883	82
H9AA	3465	898	5906	45
H42C	2905	2061	6464	53
H42D	2901	1225	6687	53
H43D	1275	1704	6283	111
H43E	1537	955	5971	111
H43F	1610	1723	5634	111
H10	2565	1429	239	96
H10E	2616	1414	248	96
H44A	856	971	-79	118
H44B	1221	1477	507	118
H45A	505	325	722	147
H45B	1610	475	1108	147
H45C	1472	-65	537	147
H44C	1006	1414	257	118
H44D	1420	824	776	118
H45D	645	518	-455	147
H45E	194	236	116	147
H45F	1243	-88	-10	147
H46A	6730	-593	3991	222
H46B	7873	-519	3889	222
H46C	7440	-1306	4036	222

H47A	6528	-1431	3128	178
H47B	6457	-569	3011	178
H48A	7957	-1500	2898	178
H48B	8103	-640	2914	178
H49A	7324	-1527	1938	178
H49B	6910	-704	1923	178
H50A	8439	-222	1871	178
H50B	9012	-975	2077	178
H51A	7746	-763	937	178
H51B	8387	-1498	1145	178
H52A	9180	-359	642	222
H52B	9869	-954	1051	222
H52C	9574	-202	1347	222
H46D	6760	-686	3609	222
H46E	7641	-197	3993	222
H46F	7883	-1010	3786	222
H47C	7218	118	2942	178
H47D	8392	-5	3197	178
H48C	7091	-836	2449	178
H48D	8136	-1159	2815	178
H49C	8017	125	2050	178
H49D	9055	-217	2405	178
H50C	8776	-1270	1842	178
H50D	7711	-968	1507	178
H51C	9537	-388	1355	178
H51D	8453	-171	989	178
H52D	8360	-1298	453	222
H52E	9447	-1527	826	222
H52F	9346	-844	370	222
H53A	9158	651	2747	222
H53B	8851	1496	2795	222
H53C	10005	1247	3003	222
H54A	10255	1108	2075	178
H54B	9091	968	1813	178
H55A	10071	2252	2056	178
H55B	8879	2195	1943	178
H56A	9986	1955	1010	178
H56B	8789	2040	931	178
H57A	8912	3274	1111	178
H57B	10111	3226	1329	178
H58A	10131	2820	284	178
H58B	9962	3683	398	178
H59A	8609	2979	-370	222
H59B	8246	3612	38	222
H59C	8189	2768	231	222
H53D	8798	1172	2341	222
H53E	9006	1687	2928	222
H53F	9931	1290	2699	222
H54C	8842	2527	2231	178
H54D	10034	2410	2354	178
H55C	8647	1966	1406	178

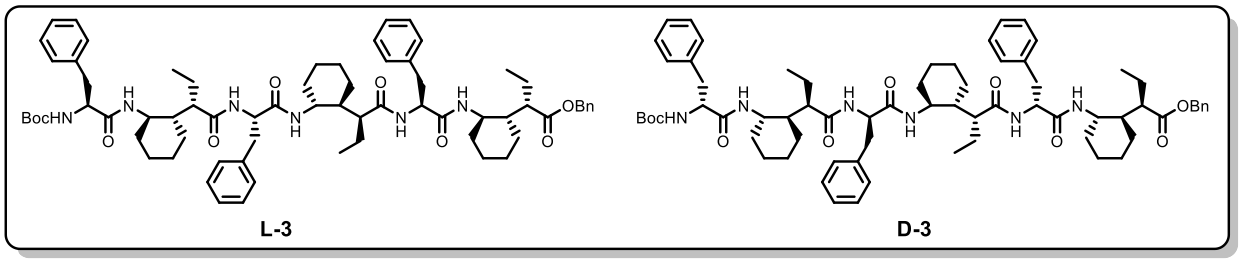
H55D	9760	1653	1536	178
H56C	9920	3082	1311	178
H56D	10206	2397	932	178
H57C	8225	2537	591	178
H57D	8645	3362	631	178
H58C	9004	2248	-165	178
H58D	9831	2894	-2	178
H59D	8391	3200	-850	222
H59E	8616	3812	-331	222
H59F	7698	3248	-343	222

Table S57. Atomic Occupancy for Gellman153.

Atom	***Occupancy***	**Atom**	***Occupancy***	**Atom**	***Occupancy***
O9	0.732(16)	H9B	0.732(16)	C42	0.732(16)
H42A	0.732(16)	H42B	0.732(16)	C43	0.732(16)
H43A	0.732(16)	H43B	0.732(16)	H43C	0.732(16)
O9A	0.268(16)	H9AA	0.268(16)	C42A	0.268(16)
H42C	0.268(16)	H42D	0.268(16)	C43A	0.268(16)
H43D	0.268(16)	H43E	0.268(16)	H43F	0.268(16)
H10	0.685(19)	H10E	0.315(19)	C44A	0.315(19)
H44A	0.315(19)	H44B	0.315(19)	C45A	0.315(19)
H45A	0.315(19)	H45B	0.315(19)	H45C	0.315(19)
C44	0.685(19)	H44C	0.685(19)	H44D	0.685(19)
C45	0.685(19)	H45D	0.685(19)	H45E	0.685(19)
H45F	0.685(19)	C46	0.5	H46A	0.5
H46B	0.5	H46C	0.5	C47	0.5
H47A	0.5	H47B	0.5	C48	0.5
H48A	0.5	H48B	0.5	C49	0.5
H49A	0.5	H49B	0.5	C50	0.5
H50A	0.5	H50B	0.5	C51	0.5
H51A	0.5	H51B	0.5	C52	0.5
H52A	0.5	H52B	0.5	H52C	0.5
C46A	0.5	H46D	0.5	H46E	0.5
H46F	0.5	C47A	0.5	H47C	0.5
H47D	0.5	C48A	0.5	H48C	0.5
H48D	0.5	C49A	0.5	H49C	0.5
H49D	0.5	C50A	0.5	H50C	0.5
H50D	0.5	C51A	0.5	H51C	0.5
H51D	0.5	C52A	0.5	H52D	0.5
H52E	0.5	H52F	0.5	C53	0.5
H53A	0.5	H53B	0.5	H53C	0.5
C54	0.5	H54A	0.5	H54B	0.5
C55	0.5	H55A	0.5	H55B	0.5
C56	0.5	H56A	0.5	H56B	0.5
C57	0.5	H57A	0.5	H57B	0.5
C58	0.5	H58A	0.5	H58B	0.5
C59	0.5	H59A	0.5	H59B	0.5
H59C	0.5	C53A	0.5	H53D	0.5

H53E	0.5	H53F	0.5	C54A	0.5
H54C	0.5	H54D	0.5	C55A	0.5
H55C	0.5	H55D	0.5	C56A	0.5
H56C	0.5	H56D	0.5	C57A	0.5
H57C	0.5	H57D	0.5	C58A	0.5
H58C	0.5	H58D	0.5	C59A	0.5
H59D	0.5	H59E	0.5	H59F	0.5

Crystallographic Experimental Section for Compound *rac*-3a



Data Collection

A colorless crystal with approximate dimensions 0.4 x 0.3 x 0.2 mm^3 was selected under oil under ambient conditions and attached to the tip of a MiTeGen MicroMount©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker SMART APEXII diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 50 frames collected at intervals of 0.5º in a 25º range about ω with the exposure time of 10 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9746 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.83 Å. A total of 136346 data were harvested by collecting 19 sets of frames with 06 scans in ω and φ with an exposure time 20-60 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were consistent for the space groups *P*2/*c* and *Pc*. The *E*-statistics strongly suggested the centrosymmetric space group *P*2/*c* that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms except those of the smallest disordered component of phenyl ring C64B were refined with anisotropic displacement coefficients. All hydrogen atoms were

included in the structure factor calculation at idealized positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

The asymmetric unit consists of one molecule of the foldamer shown in Figure 1, one fully occupied chloroform molecule, one fully occupied water molecule, as well as disordered chloroform and water molecules. Three of the phenyl groups in the foldamer exhibit positional disorder. Phenyl group C27 is disordered over two positions (major component: 51.6(15)%); phenyl group C46, over two positions (major component: 54.3(5)%); and phenyl group C64, over three positions (components: 45.0(2)%, 44.2(3)%, 10.9(3)%). Bond distance restraints and constraints as well as thermal parameter restraints were used to facilitate a chemically reasonable and computationally stable refinement.

The foldamer's conformation is stabilized by the presence of discrete hydrogen bonds between N-H and O atoms of alternating amide groups (e.g. the amide group N1 forms a hydrogen bond with amide group N3). Amide groups N4 and N6 are bridged by hydrogen bonds with the ordered water solvent molecule. This intramolecular hydrogen bonding motif is propagated in the intermolecular interactions. The penultimate amide group forms a hydrogen bond with the first amide group, and the last amide group forms a hydrogen bond with the second amide group of adjacent molecules.

A significant amount of time was invested in identifying and refining the disordered molecules chloroform and water molecules. Bond length restraints and constraints were applied to model the molecules but the resulting isotropic displacement coefficients suggested the molecules were mobile. In addition, the refinement was computationally unstable. Option SQUEEZE of program PLATON [6] was used to correct the diffraction data for diffuse scattering effects and to identify the solvate molecules. PLATON calculated the upper limit of volume that can be occupied by the solvent to be 1926 $Å^3$, or 23.3% of the unit cell volume. The program calculated 550 electrons in the unit cell for the diffuse species. The positions of two disordered, partially occupied chloroform molecules and four disordered, partially occupied water molecules were identified, but the exact composition of the disordered solvents could not be determined. It is very likely that these solvate molecules are disordered over several positions, forming hydrogen bonds. Please note that all derived results in the following tables are based on the known contents. No data are given for the diffusely scattering species.

The final least-squares refinement of 937 parameters against 14871 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0631 and 0.1764, respectively. The final difference Fourier map contained one peak ~1.8 e^-/A^3. This was in the vicinity of the disordered phenyl ring C64, and was interpreted as noise.

Summary

Crystal Data for $C_{70}\ H_{97}Cl_3N_6O_{10}$ (*M* =1288.88): monoclinic, space group *P*2/*c* (no. 13), *a* = 22.772(3) Å, *b* = 14.5752(11) Å, *c* = 25.663(3) Å, β = 103.502(13)°, *V* = 8282.1(16) $Å^3$, *Z* = 4, *T* = 100.0 K, μ(CuKα) = 1.408 mm^{-1}, *Dcalc* = 1.034 g/mm^3, 136346 reflections measured ($3.99 \leq 2\Theta \leq 136.812$), 14871 unique ($R_{int}$ = 0.0387, R_{sigma} = 0.0194) which were used in all calculations. The final R_1 was 0.0631 ($I > 2\sigma(I)$) and wR_2 was 0.1764 (all data).

References

[1] Bruker-AXS. (**2007-2014**) APEX2 (Ver. 2014.1-1), SADABS (2012-1), and SAINT+ (Ver. 8.32A) Software Reference Manuals. Bruker-AXS, Madison, Wisconsin, USA.

[2] Sheldrick, G. M. (**2008**) SHELXL. *Acta Cryst.* **A64**, 112-122.

[3] Dolomanov, O.V.; Bourhis, L.J.; Gildea, R.J.; Howard, J.A.K.; Puschmann, H. "OLEX2: a complete structure solution, refinement and analysis program". *J. Appl. Cryst.* (**2009**) **42**, 339-341.

[4] Guzei, I.A. (**2013**). Internal laboratory computer programs Gn.

[5] Guzei, I. A. (**2014**). *J. Appl. Cryst.* **47**, 806-809.

[6] A.L. Spek (**1990**) *Acta Cryst.* **A46**, C34.

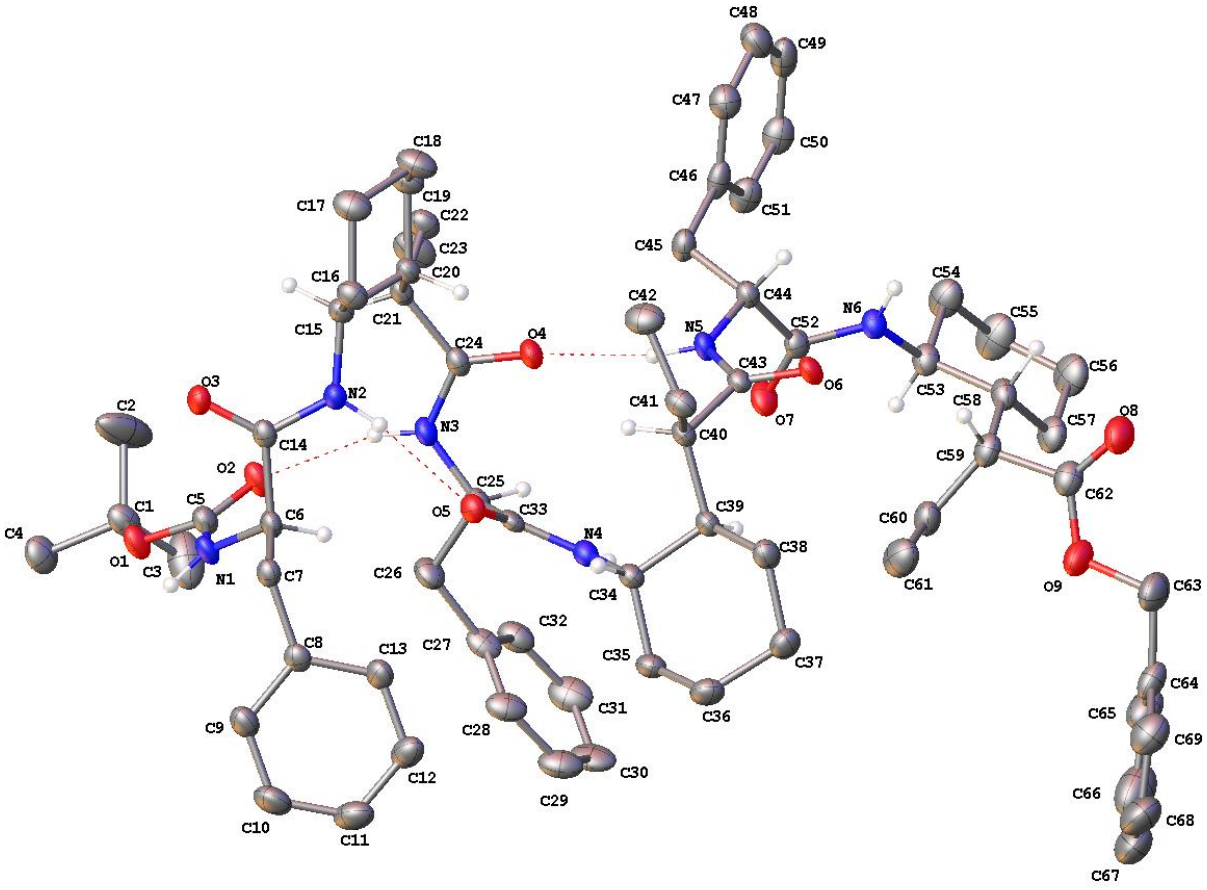


Figure S33. A molecular drawing of the foldamer of Gellman156 shown with 40% probability ellipsoids. All H atoms except those on heteroatoms and chiral carbons as well as all disordered components are omitted for clarity.

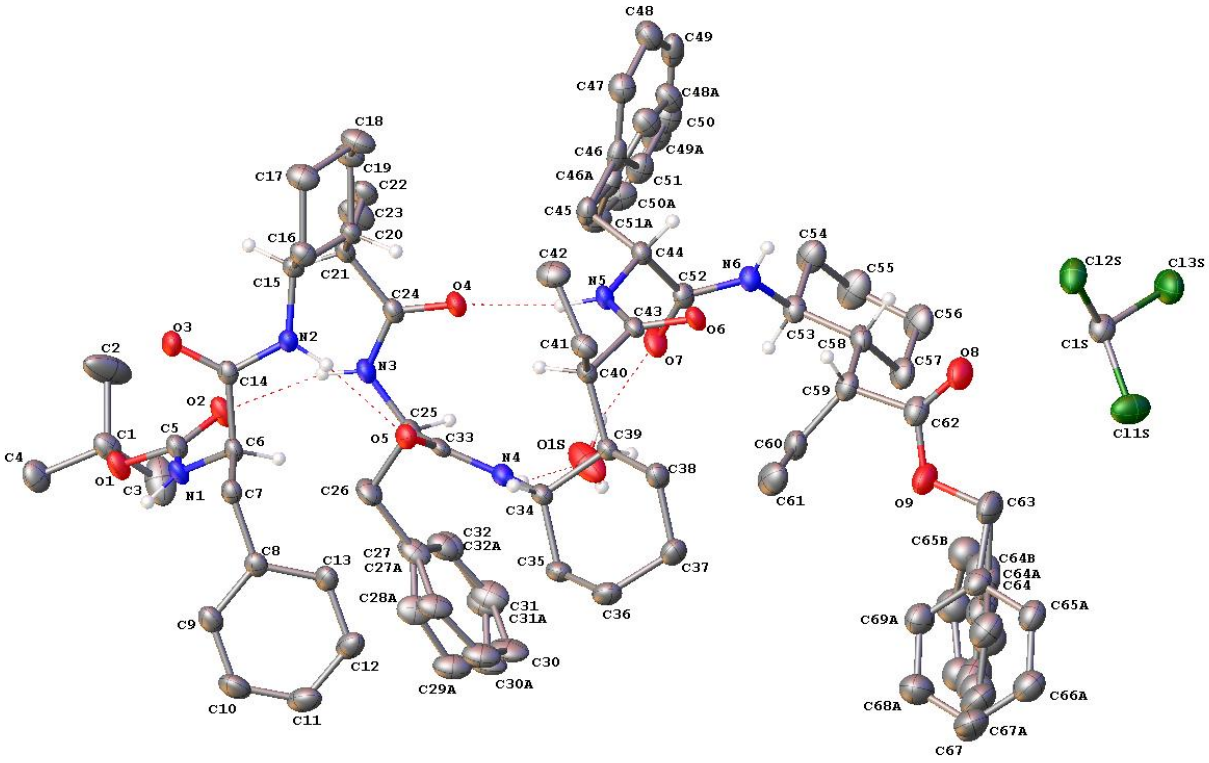


Figure S34. The asymmetric unit of Gellman156, including all disordered components. All atoms are drawn as 35% thermal probability ellipsoids. All H atoms except those on heteroatoms and chiral carbons are omitted for clarity.

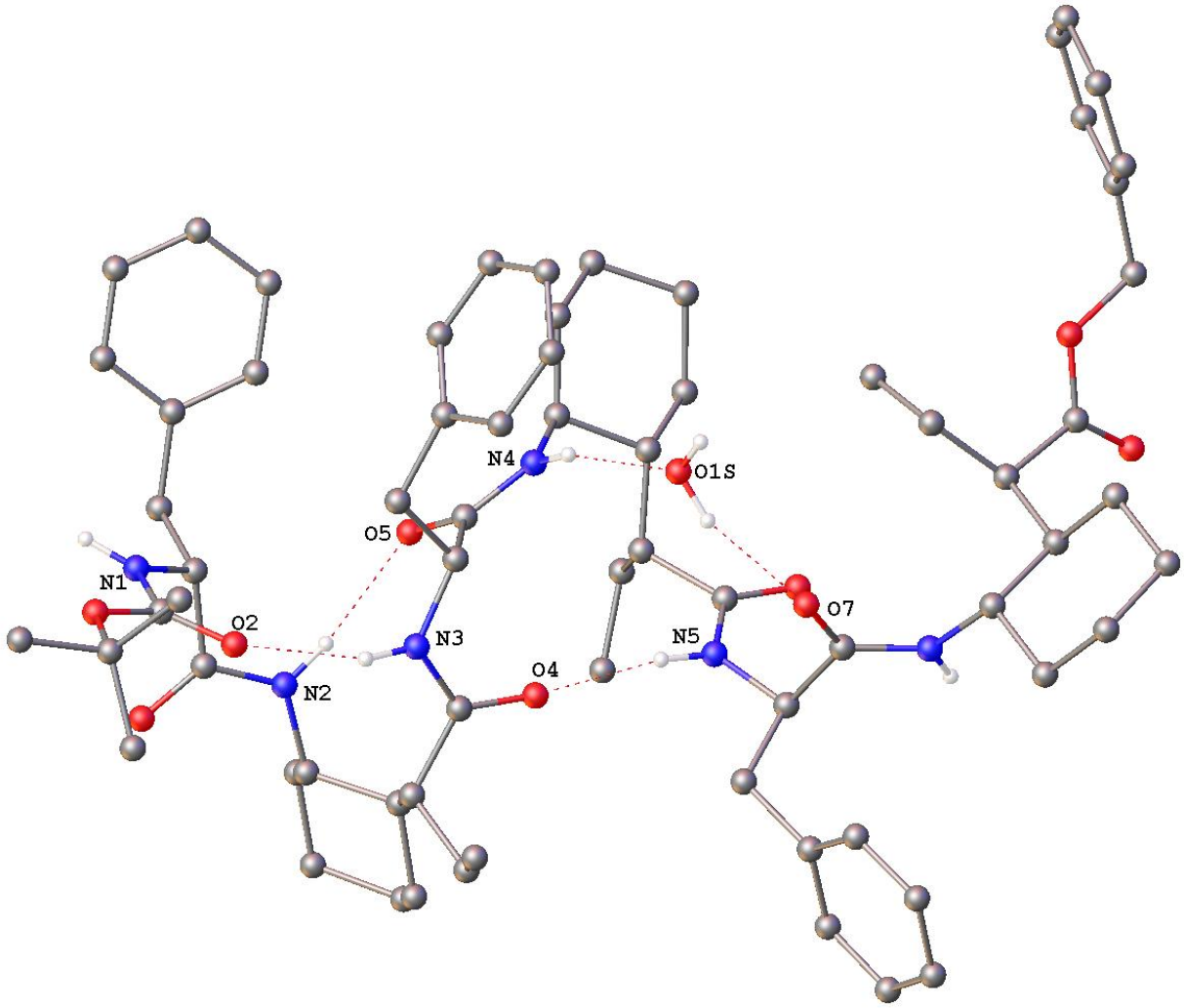


Figure S35. A packing diagram of Gellman156, illustrating the intramolecular hydrogen bonds. All H atoms except those on heteroatoms as well as disordered components are omitted for clarity.

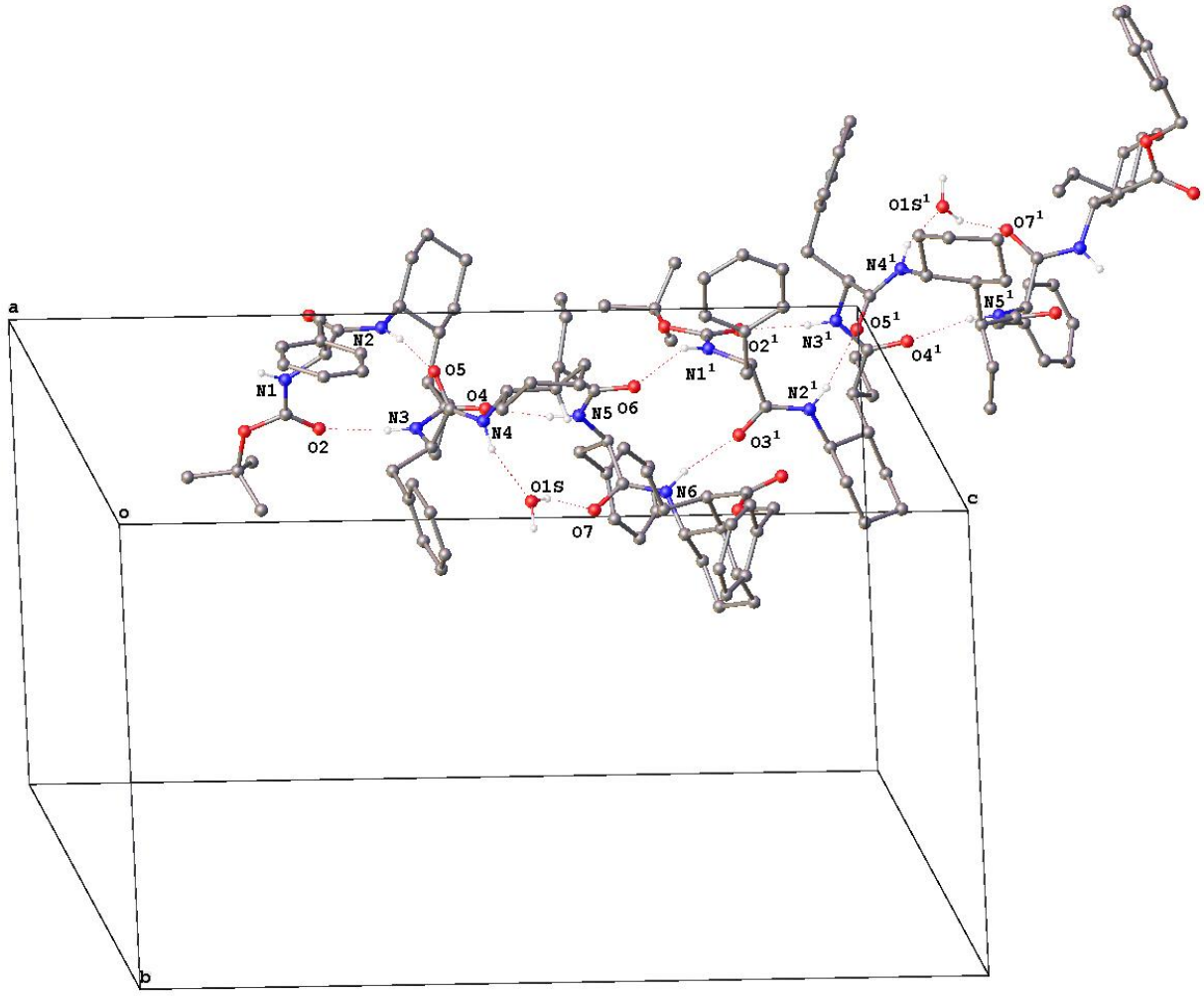


Figure S36. A packing diagram of Gellman156, illustrating the intermolecular hydrogen bonds. All H atoms except those on heteroatoms as well as disordered components are omitted for clarity.

Table S58. Crystal data and structure refinement for gellman156_sq.	
Identification code	gellman156_sq
Empirical formula	$C_{69}H_{94}N_6O_9$, $CHCl_3$, H_2O
Formula weight	1288.88
Temperature/K	100.0
Crystal system	monoclinic
Space group	*P*2/*c*
a/Å	22.772(3)
b/Å	14.5752(11)
c/Å	25.663(3)
α/°	90
β/°	103.502(13)
γ/°	90
Volume/Å^3	8282.1(16)
Z	4
ρ_{calc}mg/mm^3	1.034
μ/mm^{-1}	1.408
F(000)	2760.0
Crystal size/mm^3	0.4 × 0.3 × 0.2
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection	3.99 to 136.812°
Index ranges	$-27 \leq h \leq 26$, $-17 \leq k \leq 17$, $-30 \leq l \leq 30$
Reflections collected	136346
Independent reflections	14871 [R_{int} = 0.0387, R_{sigma} = 0.0194]
Data/restraints/parameters	14871/357/937
Goodness-of-fit on F^2	1.034
Final R indexes [I>=2σ (I)]	R_1 = 0.0631, wR_2 = 0.1680
Final R indexes [all data]	R_1 = 0.0732, wR_2 = 0.1764
Largest diff. peak/hole / e Å^{-3}	1.86/-0.61

Table S59. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for gellman156_sq. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	*x*	*y*	*z*	U(eq)
O1	6777.2(8)	1052.5(13)	2334.6(7)	35.1(4)
O2	6709.0(8)	989.9(14)	3205.3(7)	36.6(4)
O3	6905.2(8)	-1357.4(13)	3171.2(7)	32.6(4)
O4	6780.9(8)	650.2(13)	5185.7(6)	33.7(4)
O5	8003.3(8)	376.3(12)	4745.4(7)	30.7(4)
O6	7887.2(8)	721.3(12)	7080.8(6)	30.5(4)
O7	6938.4(10)	2934.1(13)	6435.1(7)	40.2(5)
O8	8499.1(10)	2970.7(15)	8842.4(8)	51.3(5)
O9	8967.2(10)	3892.2(15)	8369.2(8)	48.3(5)
N1	7458.6(9)	260.6(15)	2923.2(8)	28.1(4)
N2	7176.9(9)	-955.5(14)	4047.7(8)	26.3(4)
N3	6863(1)	1083.4(14)	4365.9(8)	29.1(4)

N4	8237.2(9)	1563.4(14)	5326.9(8)	27.8(4)
N5	7172.2(9)	1005.2(14)	6327.6(8)	27.5(4)
N6	6880.4(9)	2552.3(14)	7278.8(8)	28.7(4)
C1	6260.9(12)	1673(2)	2169.3(11)	38.0(6)
C2	5701.5(16)	1234(4)	2276.0(19)	91.0(17)
C3	6399(2)	2583(3)	2445.2(15)	77.5(13)
C4	6212.4(14)	1767(2)	1571.5(11)	46.2(7)
C5	6960.2(11)	779.5(18)	2850.5(10)	30.4(5)
C6	7674.1(10)	-191.1(16)	3437.3(9)	25.3(5)
C7	8288.2(11)	-649.7(17)	3460.9(9)	27.2(5)
C8	8797.6(11)	27.4(17)	3519.4(10)	28.7(5)
C9	9042.2(12)	259(2)	3091.4(11)	38.6(6)
C10	9529.7(14)	855(2)	3159.1(13)	50.0(8)
C11	9774.7(14)	1236(2)	3654.2(15)	52.1(8)
C12	9529.9(13)	1023(2)	4083.4(13)	44.3(7)
C13	9044.4(12)	429.4(19)	4016.0(11)	34.8(6)
C14	7212.2(10)	-894.4(16)	3537.2(9)	25.9(5)
C15	6764.3(11)	-1591.1(17)	4222.2(10)	27.6(5)
C16	7085.5(12)	-2486.4(18)	4432.2(11)	34.1(6)
C17	6642.6(14)	-3170(2)	4573.8(13)	43.0(7)
C18	6327.8(15)	-2753(2)	4977.8(13)	46.1(7)
C19	6019.4(13)	-1847.5(19)	4776.0(12)	37.2(6)
C20	6465.6(11)	-1151.5(17)	4640.3(9)	27.5(5)
C21	6185.5(11)	-197.0(18)	4450.1(9)	28.4(5)
C22	5588.3(12)	25(2)	4603.5(11)	36.4(6)
C23	5342.1(14)	963(2)	4395.3(14)	51.8(8)
C24	6633.1(11)	545.1(17)	4694.6(9)	28.0(5)
C25	7352.4(12)	1706.0(17)	4594.7(9)	29.5(5)
C26	7519.3(14)	2282.8(19)	4147.9(11)	39.5(6)
C27	7875(4)	3149(4)	4341(4)	36.8(18)
C28	8480(4)	3141(3)	4322(4)	44.6(17)
C29	8836(3)	3912(4)	4478(5)	51.4(17)
C30	8588(3)	4692(4)	4653(4)	53.9(18)
C31	7983(3)	4701(5)	4671(4)	52.4(18)
C32	7626(3)	3929(6)	4515(5)	43.2(18)
C27A	7942(4)	3076(5)	4372(4)	34.8(18)
C28A	8569(4)	3049(4)	4497(4)	42.8(17)
C29A	8899(3)	3803(5)	4735(4)	50.6(17)
C30A	8602(3)	4585(4)	4847(4)	51.4(18)
C31A	7975(3)	4612(5)	4723(4)	52.9(19)
C32A	7644(3)	3858(7)	4485(5)	41.2(18)
C33	7896.5(11)	1145.4(17)	4901.0(9)	27.0(5)
C34	8775.3(11)	1169.6(17)	5685.3(9)	28.1(5)
C35	9318.0(12)	1790(2)	5685.8(11)	36.6(6)
C36	9878.1(12)	1487(2)	6100.3(12)	41.6(7)
C37	9748.2(12)	1484(2)	6657.8(11)	39.6(6)
C38	9228.9(12)	829(2)	6667.8(10)	34.8(6)
C39	8652.4(11)	1067.0(17)	6248.7(9)	26.9(5)
C40	8121.4(11)	382.7(17)	6227.2(9)	27.0(5)
C41	8309.4(12)	-613.3(18)	6361.9(11)	34.6(6)
C42	7761.1(14)	-1251(2)	6246.9(14)	47.0(7)

C43	7720.8(11)	712.4(16)	6586.5(9)	25.1(5)
C44	6726.0(11)	1359.4(17)	6598.0(9)	26.5(5)
C45	6103.9(12)	1313.0(19)	6191.8(10)	33.4(6)
C46	5571(2)	1543(6)	6432(3)	36.1(16)
C47	5244(2)	869(4)	6609(2)	42.8(14)
C48	4754(3)	1056(4)	6831(2)	49.0(15)
C49	4589(3)	1943(4)	6884(2)	46.4(15)
C50	4899(6)	2649(6)	6706(7)	43(3)
C51	5382(2)	2449(4)	6480(2)	41.5(14)
C46A	5632(3)	1955(5)	6311(3)	32.8(18)
C47A	5292(3)	1720(5)	6664(2)	40.5(17)
C48A	4865(7)	2334(8)	6772(8)	41(3)
C49A	4790(3)	3174(6)	6530(3)	47.0(18)
C50A	5123(3)	3421(4)	6165(3)	46.9(18)
C51A	5543(2)	2808(4)	6052(2)	39.8(16)
C52	6865.0(11)	2350.6(17)	6770.1(9)	28.7(5)
C53	6934.2(12)	3491.6(17)	7485(1)	33.1(6)
C54	6343.5(14)	3807(2)	7606.7(14)	49.8(8)
C55	6385.6(16)	4808(3)	7786.8(16)	61.9(10)
C56	6910.6(16)	4958(2)	8262.6(14)	55.1(8)
C57	7502.2(14)	4612.1(19)	8150.9(12)	43.0(7)
C58	7455.0(13)	3599.0(18)	7983.6(10)	34.0(6)
C59	8054.8(13)	3194.0(19)	7901.6(11)	36.5(6)
C60	8260.0(13)	3533(2)	7407.6(11)	41.2(6)
C61	8769.5(16)	2944(3)	7289.5(15)	57.2(9)
C62	8524.3(14)	3324.0(19)	8423.1(11)	39.5(6)
C63	9438.4(16)	4078(3)	8844.1(13)	57.7(9)
C64	9997.8(16)	4341(3)	8665.8(17)	41.5(15)
C65	10520(2)	4422(3)	9071.1(13)	46.9(15)
C66	11045.4(17)	4764(4)	8956.1(16)	58.9(17)
C67	11048.6(17)	5025(5)	8435.9(18)	71(2)
C68	10526(2)	4944(4)	8030.6(14)	67.5(18)
C69	10001.0(16)	4602(4)	8145.6(15)	52.4(16)
C64A	9876(2)	4747(3)	8691(2)	43.6(16)
C65A	9685(2)	5618(3)	8505(2)	63(2)
C66A	10076(3)	6197(3)	8317(3)	95(3)
C67A	10656(3)	5905(5)	8316(3)	84(2)
C68A	10847(2)	5034(5)	8502(3)	79(2)
C69A	10457(2)	4455(3)	8689(3)	63.1(17)
C64B	9659(10)	4948(10)	8632(10)	63(4)
C65B	9143(8)	5464(14)	8428(10)	63(4)
C66B	9189(9)	6306(13)	8186(9)	63(4)
C67B	9751(11)	6633(10)	8149(10)	63(4)
C68B	10267(8)	6118(12)	8353(10)	63(4)
C69B	10221(8)	5275(11)	8595(10)	63(4)
Cl1S	9569.2(4)	2953.1(6)	10187.1(4)	61.5(2)
Cl2S	8714.4(4)	1435.8(5)	10021.0(3)	51.3(2)
Cl3S	8466.0(4)	3080.1(6)	10549.5(3)	60.6(2)
C1S	8802.1(14)	2631(2)	10053.2(12)	40.9(6)
O1S	7762.8(17)	3096.0(16)	5793.6(11)	74.3(8)

Table S60. Anisotropic Displacement Parameters ($Å^2 \times 10^3$) for gellman156_sq. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O1	34.7(9)	49.3(11)	22.3(8)	7.2(8)	8.4(7)	11.1(8)
O2	37.6(10)	49.9(11)	25.1(9)	3.7(8)	12.6(8)	13.9(8)
O3	32.8(9)	41.6(10)	24.2(8)	-8.0(7)	8.2(7)	-3.1(8)
O4	40.9(10)	42(1)	19.8(8)	-2.2(7)	10.4(7)	-5.2(8)
O5	34.8(9)	33.1(9)	26.1(8)	-5.3(7)	10.9(7)	-0.6(7)
O6	30.5(9)	43.7(10)	18.3(8)	3.3(7)	7.9(7)	1.1(7)
O7	59.0(12)	35.2(10)	24.5(9)	6.5(7)	6.1(8)	-1.7(9)
O8	60.6(14)	54.5(13)	32.9(11)	4.9(9)	-0.8(10)	-8.4(10)
O9	52.8(12)	50.0(12)	34.4(10)	1.1(9)	-5.3(9)	-10.9(10)
N1	28.5(10)	39.3(12)	19.4(9)	1.3(8)	11.2(8)	5.1(9)
N2	25.5(10)	33.3(11)	20.9(9)	-3.3(8)	6.7(8)	-1.9(8)
N3	35.5(11)	34.2(11)	18.2(9)	1.7(8)	7.1(8)	0.3(9)
N4	35.2(11)	25.8(10)	24.4(10)	-0.1(8)	10.8(9)	-3.5(8)
N5	30.4(10)	35.9(11)	16.7(9)	-0.9(8)	6.8(8)	2.0(9)
N6	35.5(11)	26.5(10)	23.7(10)	0.3(8)	6.2(9)	2.0(8)
C1	34.6(14)	46.2(16)	34.1(14)	11.8(12)	9.8(12)	11.0(12)
C2	33.4(18)	151(5)	88(3)	64(3)	13.1(19)	6(2)
C3	129(4)	50(2)	50(2)	4.0(17)	12(2)	27(2)
C4	46.9(17)	55.6(18)	33.3(14)	12.9(13)	4.0(13)	11.1(14)
C5	31.1(13)	38.1(14)	22.9(12)	2(1)	8.1(10)	1.9(10)
C6	27.2(12)	31.3(12)	18.4(10)	-2.3(9)	7.3(9)	2.2(9)
C7	27.1(12)	32.3(13)	22.8(11)	-2.5(9)	6.6(10)	3.4(10)
C8	24.4(11)	34.5(13)	28.5(12)	-1.2(10)	8.8(10)	5.3(10)
C9	34.7(14)	52.8(17)	30.9(13)	-0.9(12)	12.6(12)	2.0(12)
C10	43.3(16)	64(2)	51.1(18)	3.5(15)	27.4(15)	-4.4(15)
C11	38.5(16)	53.8(19)	69(2)	-9.5(16)	23.5(16)	-11.2(14)
C12	32.3(14)	51.6(18)	50.5(17)	-16.5(14)	13.0(13)	-5.9(12)
C13	31.8(13)	44.3(15)	31.2(13)	-7.6(11)	13.1(11)	-0.2(11)
C14	25.0(11)	30.6(12)	22.8(11)	-1.5(9)	7.2(10)	6.0(9)
C15	25.3(12)	33.7(13)	25.4(12)	-1.2(10)	8.9(10)	-2(1)
C16	32.1(13)	37.5(14)	35.2(13)	2.9(11)	12.8(11)	4.2(11)
C17	46.8(16)	33.3(14)	53.1(17)	8.4(13)	20.2(14)	3.2(12)
C18	49.4(17)	41.5(16)	55.1(18)	11.7(14)	28.0(15)	1.3(13)
C19	36.7(14)	38.0(14)	42.2(15)	2.9(12)	20.0(12)	-0.2(11)
C20	25.7(11)	35.8(13)	22.0(11)	1.3(10)	7.8(10)	0(1)
C21	26.8(12)	39.4(14)	20.6(11)	-0.2(10)	8.7(10)	1.7(10)
C22	29.9(13)	45.4(16)	36.8(14)	1.2(12)	13.8(11)	4.0(11)
C23	42.4(16)	59(2)	61(2)	12.0(16)	25.6(15)	20.5(15)
C24	29.2(12)	34.6(13)	21.6(11)	0.3(10)	8.6(10)	4.5(10)
C25	38.2(14)	29.7(13)	22.8(11)	1.1(10)	11.5(11)	-0.5(10)
C26	53.5(17)	36.6(15)	30.8(13)	8.1(11)	14.5(13)	-2.1(12)
C27	38(3)	35(3)	39(3)	18(3)	13(3)	4(3)
C28	44(3)	29(3)	62(4)	17(3)	13(3)	14(2)
C29	47(3)	40(3)	71(4)	15(3)	22(3)	-1(2)
C30	64(3)	35(3)	72(4)	14(3)	35(3)	-7(3)
C31	63(3)	30(3)	71(3)	8(3)	30(3)	2(3)

C32	50(3)	36(3)	48(3)	6(3)	20(3)	8(3)
C27A	45(3)	24(3)	40(3)	13(3)	18(3)	10(3)
C28A	42(3)	33(3)	57(4)	18(3)	19(3)	5(3)
C29A	49(3)	38(3)	71(4)	16(3)	26(3)	-1(3)
C30A	60(3)	28(3)	75(4)	15(3)	33(3)	-5(2)
C31A	65(3)	29(3)	72(4)	9(3)	30(3)	8(3)
C32A	46(3)	36(3)	47(3)	9(3)	22(3)	7(3)
C33	32.0(12)	31.3(13)	21.9(11)	3.2(10)	14.7(10)	-4.9(10)
C34	30.9(12)	31.5(13)	24.3(12)	0.1(10)	11(1)	-4.1(10)
C35	35.5(14)	42.3(15)	35.4(14)	-0.3(12)	15.2(12)	-11.4(12)
C36	30.0(13)	49.1(17)	48.1(16)	-2.8(13)	13.7(13)	-7.6(12)
C37	29.6(13)	49.9(17)	37.1(15)	-3.3(12)	3.4(12)	-3.6(12)
C38	30.9(13)	45.4(15)	28.1(12)	1.5(11)	7.0(11)	0.8(11)
C39	28.6(12)	31.4(12)	22.6(11)	-1.2(9)	9.7(10)	-1.3(10)
C40	30.5(12)	31.3(13)	21.1(11)	0.8(9)	10(1)	-1.5(10)
C41	39.0(14)	32.1(13)	36.5(14)	2.4(11)	16.7(12)	2.2(11)
C42	50.8(17)	31.4(14)	63(2)	0.1(13)	21.2(15)	-4.1(13)
C43	30.9(12)	24.4(11)	21.5(11)	1.6(9)	9.6(10)	-3.1(9)
C44	28.1(12)	30.6(12)	21.5(11)	-0.5(9)	7(1)	1.2(10)
C45	32.3(13)	40.6(14)	25.7(12)	-7.2(11)	3.3(11)	2.9(11)
C46	32(3)	43(4)	27(3)	-11(3)	-6(3)	11(3)
C47	45(3)	44(3)	39(3)	-1(2)	9(2)	3(2)
C48	50(3)	63(4)	36(3)	3(3)	13(3)	-1(3)
C49	37(3)	68(4)	32(3)	-12(3)	2(3)	8(3)
C50	39(4)	44(7)	42(6)	-4(6)	2(3)	11(5)
C51	34(3)	44(3)	43(3)	-4(3)	3(2)	7(2)
C46A	28(3)	33(4)	32(4)	-10(3)	-4(3)	3(3)
C47A	35(3)	47(4)	38(3)	5(3)	7(3)	8(3)
C48A	36(5)	54(9)	33(5)	7(7)	10(4)	10(7)
C49A	38(4)	50(5)	50(4)	-6(4)	4(3)	8(3)
C50A	36(3)	41(4)	63(4)	6(3)	9(3)	8(3)
C51A	29(3)	41(3)	46(4)	8(3)	4(3)	0(2)
C52	29.3(12)	32.4(13)	22.7(11)	2.3(10)	2.9(10)	3.7(10)
C53	41.4(14)	27.2(13)	30.0(13)	-1.1(10)	6.6(11)	2.5(11)
C54	43.1(16)	51.3(18)	54.6(18)	-15.3(15)	10.6(15)	6.6(14)
C55	56(2)	53(2)	74(2)	-21.8(18)	11.3(18)	16.5(16)
C56	64(2)	44.7(18)	58(2)	-19.7(15)	15.7(17)	4.4(15)
C57	55.2(18)	34.3(15)	39.2(15)	-10.8(12)	10.3(13)	-0.9(13)
C58	46.1(15)	29.9(13)	26.2(12)	-2.8(10)	8.9(11)	-1.5(11)
C59	41.1(15)	32.4(13)	33.0(14)	-6.5(11)	2.9(12)	-0.6(11)
C60	40.5(15)	50.1(17)	31.3(14)	-8.8(12)	5.1(12)	-1.5(13)
C61	53.9(19)	61(2)	60(2)	-23.4(17)	18.9(17)	-3.6(16)
C62	46.4(16)	33.3(14)	35.3(15)	-1.0(12)	2.5(13)	0.7(12)
C63	58(2)	69(2)	37.3(16)	1.4(15)	-6.1(15)	-13.1(17)
C64	47(3)	32(3)	42(3)	0(3)	3(3)	-7(3)
C65	53(3)	43(3)	42(3)	-4(3)	4(3)	-5(3)
C66	53(3)	60(4)	61(4)	-9(3)	10(3)	-10(3)
C67	57(4)	88(4)	70(4)	-10(4)	17(4)	-31(4)
C68	59(4)	85(4)	61(4)	1(3)	20(3)	-15(3)
C69	49(3)	58(3)	46(3)	2(3)	3(3)	-5(3)
C64A	45(3)	38(3)	38(3)	-5(3)	-9(3)	-2(3)

C65A	72(5)	50(4)	57(4)	0(3)	-4(4)	-20(3)
C66A	113(6)	84(5)	70(5)	3(4)	-15(5)	-22(5)
C67A	85(4)	99(5)	64(4)	-20(4)	11(4)	-43(4)
C68A	62(4)	95(4)	76(4)	-21(4)	11(4)	-32(4)
C69A	61(3)	57(3)	66(3)	-10(3)	6(3)	-7(3)
Cl1S	41.8(4)	63.6(5)	73.2(6)	23.3(4)	1.3(4)	-1.4(4)
Cl2S	74.9(5)	33.9(4)	40.6(4)	-0.1(3)	4.5(4)	0.4(3)
Cl3S	77.0(6)	55.8(5)	52.9(5)	-15.9(4)	22.9(4)	-9.0(4)
C1S	47.4(16)	36.6(15)	36.8(14)	2.7(12)	5.9(13)	-0.4(12)
O1S	133(3)	37.0(12)	68.7(17)	4.6(12)	56.1(18)	7.1(14)

Table S61. Bond Lengths for gellman156_sq.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
O1	C1	1.465(3)	C34	C35	1.531(3)
O1	C5	1.352(3)	C34	C39	1.543(3)
O2	C5	1.222(3)	C35	C36	1.523(4)
O3	C14	1.233(3)	C36	C37	1.527(4)
O4	C24	1.236(3)	C37	C38	1.524(4)
O5	C33	1.233(3)	C38	C39	1.530(3)
O6	C43	1.236(3)	C39	C40	1.558(3)
O7	C52	1.248(3)	C40	C41	1.530(3)
O8	C62	1.206(4)	C40	C43	1.519(3)
O9	C62	1.337(4)	C41	C42	1.529(4)
O9	C63	1.449(4)	C44	C45	1.552(3)
N1	C5	1.340(3)	C44	C52	1.522(3)
N1	C6	1.453(3)	C45	C46	1.522(5)
N2	C14	1.334(3)	C45	C46A	1.510(5)
N2	C15	1.462(3)	C46	C47	1.373(8)
N3	C24	1.344(3)	C46	C51	1.402(8)
N3	C25	1.451(3)	C47	C48	1.393(7)
N4	C33	1.331(3)	C48	C49	1.363(8)
N4	C34	1.467(3)	C49	C50	1.385(8)
N5	C43	1.340(3)	C50	C51	1.389(8)
N5	C44	1.453(3)	C46A	C47A	1.366(9)
N6	C52	1.331(3)	C46A	C51A	1.401(8)
N6	C53	1.463(3)	C47A	C48A	1.396(8)
C1	C2	1.507(5)	C48A	C49A	1.365(8)
C1	C3	1.502(5)	C49A	C50A	1.384(8)
C1	C4	1.518(4)	C50A	C51A	1.388(8)
C6	C7	1.538(3)	C53	C54	1.521(4)
C6	C14	1.532(3)	C53	C58	1.536(4)
C7	C8	1.504(3)	C54	C55	1.526(5)
C8	C9	1.385(4)	C55	C56	1.512(5)
C8	C13	1.396(4)	C56	C57	1.527(5)
C9	C10	1.388(4)	C57	C58	1.535(4)
C10	C11	1.380(5)	C58	C59	1.547(4)
C11	C12	1.381(5)	C59	C60	1.532(4)
C12	C13	1.383(4)	C59	C62	1.517(4)

C15	C16	1.531(4)	C60	C61	1.530(4)
C15	C20	1.538(3)	C63	C64	1.500(5)
C16	C17	1.521(4)	C63	C64A	1.510(5)
C17	C18	1.518(4)	C63	C64B	1.512(6)
C18	C19	1.528(4)	C64	C65	1.3900
C19	C20	1.532(4)	C64	C69	1.3900
C20	C21	1.560(3)	C65	C66	1.3900
C21	C22	1.536(3)	C66	C67	1.3900
C21	C24	1.518(4)	C67	C68	1.3900
C22	C23	1.525(4)	C68	C69	1.3900
C25	C26	1.539(3)	C64A	C65A	1.3900
C25	C33	1.538(4)	C64A	C69A	1.3900
C26	C27	1.519(4)	C65A	C66A	1.3900
C26	C27A	1.528(4)	C66A	C67A	1.3900
C27	C28	1.3900	C67A	C68A	1.3900
C27	C32	1.3900	C68A	C69A	1.3900
C28	C29	1.3900	C64B	C65B	1.3900
C29	C30	1.3900	C64B	C69B	1.3900
C30	C31	1.3900	C65B	C66B	1.3900
C31	C32	1.3900	C66B	C67B	1.3900
C27A	C28A	1.3900	C67B	C68B	1.3900
C27A	C32A	1.3900	C68B	C69B	1.3900
C28A	C29A	1.3900	Cl1S	C1S	1.763(3)
C29A	C30A	1.3900	Cl2S	C1S	1.754(3)
C30A	C31A	1.3900	Cl3S	C1S	1.758(3)
C31A	C32A	1.3900			

Table S62. Bond Angles for gellman156_sq.

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C5	O1	C1	120.6(2)	C38	C39	C34	111.5(2)
C62	O9	C63	117.2(2)	C38	C39	C40	114.3(2)
C5	N1	C6	118.5(2)	C41	C40	C39	115.2(2)
C14	N2	C15	122.2(2)	C43	C40	C39	110.31(19)
C24	N3	C25	119.1(2)	C43	C40	C41	109.82(19)
C33	N4	C34	125.3(2)	C42	C41	C40	111.1(2)
C43	N5	C44	123.45(19)	O6	C43	N5	122.2(2)
C52	N6	C53	122.9(2)	O6	C43	C40	122.9(2)
O1	C1	C2	109.9(3)	N5	C43	C40	115.0(2)
O1	C1	C3	110.0(3)	N5	C44	C45	107.11(19)
O1	C1	C4	102.3(2)	N5	C44	C52	110.8(2)
C2	C1	C4	110.9(3)	C52	C44	C45	109.4(2)
C3	C1	C2	112.5(4)	C46	C45	C44	114.1(3)
C3	C1	C4	110.7(3)	C46A	C45	C44	114.8(3)
O2	C5	O1	124.4(2)	C47	C46	C45	121.5(6)
O2	C5	N1	124.5(2)	C47	C46	C51	116.3(4)
N1	C5	O1	111.1(2)	C51	C46	C45	122.2(6)
N1	C6	C7	110.10(19)	C46	C47	C48	122.9(5)
N1	C6	C14	110.82(19)	C49	C48	C47	119.5(6)

C14	C6	C7	110.85(19)	C48	C49	C50	119.9(6)
C8	C7	C6	113.1(2)	C49	C50	C51	119.7(7)
C9	C8	C7	121.7(2)	C50	C51	C46	121.6(6)
C9	C8	C13	118.1(2)	C47A	C46A	C45	121.3(6)
C13	C8	C7	120.2(2)	C47A	C46A	C51A	119.7(5)
C8	C9	C10	120.8(3)	C51A	C46A	C45	119.0(6)
C11	C10	C9	120.4(3)	C46A	C47A	C48A	120.0(6)
C10	C11	C12	119.5(3)	C49A	C48A	C47A	120.2(8)
C11	C12	C13	120.0(3)	C48A	C49A	C50A	120.8(7)
C12	C13	C8	121.1(3)	C49A	C50A	C51A	119.1(6)
O3	C14	N2	123.9(2)	C50A	C51A	C46A	120.2(6)
O3	C14	C6	121.9(2)	O7	C52	N6	123.2(2)
N2	C14	C6	114.2(2)	O7	C52	C44	119.9(2)
N2	C15	C16	111.1(2)	N6	C52	C44	116.8(2)
N2	C15	C20	111.7(2)	N6	C53	C54	110.7(2)
C16	C15	C20	111.2(2)	N6	C53	C58	112.3(2)
C17	C16	C15	110.7(2)	C54	C53	C58	110.2(2)
C18	C17	C16	110.3(2)	C53	C54	C55	110.8(3)
C17	C18	C19	111.5(2)	C56	C55	C54	111.4(3)
C18	C19	C20	111.9(2)	C55	C56	C57	111.5(3)
C15	C20	C21	111.61(19)	C56	C57	C58	111.3(3)
C19	C20	C15	107.9(2)	C53	C58	C59	112.5(2)
C19	C20	C21	114.6(2)	C57	C58	C53	108.7(2)
C22	C21	C20	115.4(2)	C57	C58	C59	113.5(2)
C24	C21	C20	108.67(19)	C60	C59	C58	116.0(2)
C24	C21	C22	107.0(2)	C62	C59	C58	107.2(2)
C23	C22	C21	112.0(2)	C62	C59	C60	113.7(2)
O4	C24	N3	120.9(2)	C61	C60	C59	112.0(3)
O4	C24	C21	120.4(2)	O8	C62	O9	123.4(3)
N3	C24	C21	118.6(2)	O8	C62	C59	124.3(3)
N3	C25	C26	109.9(2)	O9	C62	C59	112.3(2)
N3	C25	C33	109.0(2)	O9	C63	C64	107.8(3)
C33	C25	C26	111.2(2)	O9	C63	C64A	107.8(3)
C27	C26	C25	114.6(5)	O9	C63	C64B	95.5(10)
C27A	C26	C25	112.1(5)	C65	C64	C63	115.6(3)
C28	C27	C26	116.0(6)	C65	C64	C69	120.0
C28	C27	C32	120.0	C69	C64	C63	123.9(3)
C32	C27	C26	124.0(6)	C64	C65	C66	120.0
C27	C28	C29	120.0	C67	C66	C65	120.0
C28	C29	C30	120.0	C66	C67	C68	120.0
C31	C30	C29	120.0	C67	C68	C69	120.0
C30	C31	C32	120.0	C68	C69	C64	120.0
C31	C32	C27	120.0	C65A	C64A	C63	120.5(4)
C28A	C27A	C26	126.0(6)	C65A	C64A	C69A	120.0
C28A	C27A	C32A	120.0	C69A	C64A	C63	119.3(4)
C32A	C27A	C26	113.9(7)	C64A	C65A	C66A	120.0
C29A	C28A	C27A	120.0	C65A	C66A	C67A	120.0
C28A	C29A	C30A	120.0	C68A	C67A	C66A	120.0
C31A	C30A	C29A	120.0	C67A	C68A	C69A	120.0
C32A	C31A	C30A	120.0	C68A	C69A	C64A	120.0
C31A	C32A	C27A	120.0	C65B	C64B	C63	105.5(14)

O5	C33	N4	124.1(2)	C65B	C64B	C69B	120.0
O5	C33	C25	120.9(2)	C69B	C64B	C63	134.5(14)
N4	C33	C25	115.0(2)	C66B	C65B	C64B	120.0
N4	C34	C35	109.0(2)	C65B	C66B	C67B	120.0
N4	C34	C39	108.53(19)	C68B	C67B	C66B	120.0
C35	C34	C39	112.3(2)	C67B	C68B	C69B	120.0
C36	C35	C34	112.0(2)	C68B	C69B	C64B	120.0
C35	C36	C37	110.0(2)	Cl2S	C1S	Cl1S	111.72(17)
C38	C37	C36	109.8(2)	Cl2S	C1S	Cl3S	109.86(16)
C37	C38	C39	112.8(2)	Cl3S	C1S	Cl1S	109.80(16)
C34	C39	C40	109.74(19)				

Table S63. Hydrogen Bonds for gellman156_sq.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
N1	H1	O6[1]	0.88	2.17	2.943(3)	146.6
N2	H2	O5	0.88	2.12	2.990(3)	169.7
N3	H3	O2	0.88	2.08	2.920(3)	159.3
N4	H4	O1S	0.88	2.04	2.863(3)	156.2
N5	H5	O4	0.88	2.04	2.903(3)	167.3
N6	H6	O3[2]	0.88	2.01	2.867(3)	162.9
C1S	H1S	O8	1.00	2.16	3.062(4)	148.7
O1S	H1SA	O7	0.87	1.95	2.781(3)	158.9

[1]+X,-Y,-1/2+Z; [2]+X,-Y,1/2+Z

Table S64. Torsion Angles for gellman156_sq.

A	B	C	D	Angle/°	A	B	C	D	Angle/°
O9	C63	C64	C65	-171.5(3)	C35	C34	C39	C40	-176.5(2)
O9	C63	C64	C69	16.2(5)	C35	C36	C37	C38	59.5(3)
O9	C63	C64A	C65A	62.1(5)	C36	C37	C38	C39	-57.3(3)
O9	C63	C64A	C69A	-111.9(4)	C37	C38	C39	C34	51.8(3)
O9	C63	C64B	C65B	48.4(11)	C37	C38	C39	C40	177.0(2)
O9	C63	C64B	C69B	-129.5(19)	C38	C39	C40	C41	-32.3(3)
N1	C6	C7	C8	73.0(2)	C38	C39	C40	C43	92.7(2)
N1	C6	C14	O3	36.0(3)	C39	C34	C35	C36	52.8(3)
N1	C6	C14	N2	-144.1(2)	C39	C40	C41	C42	-171.3(2)
N2	C15	C16	C17	-175.8(2)	C39	C40	C43	O6	-69.1(3)
N2	C15	C20	C19	177.1(2)	C39	C40	C43	N5	110.2(2)
N2	C15	C20	C21	50.3(3)	C41	C40	C43	O6	58.9(3)
N3	C25	C26	C27	161.5(5)	C41	C40	C43	N5	-121.9(2)
N3	C25	C26	C27A	168.7(5)	C43	N5	C44	C45	-161.4(2)
N3	C25	C33	O5	33.6(3)	C43	N5	C44	C52	79.4(3)
N3	C25	C33	N4	-148.0(2)	C43	C40	C41	C42	63.5(3)
N4	C34	C35	C36	173.1(2)	C44	N5	C43	O6	0.2(4)
N4	C34	C39	C38	-169.4(2)	C44	N5	C43	C40	-179.1(2)
N4	C34	C39	C40	62.9(2)	C44	C45	C46	C47	-95.6(6)

N5	C44	C45	C46	172.5(4)	C44	C45	C46	C51	84.9(6)
N5	C44	C45	C46A	-158.0(4)	C44	C45	C46A	C47A	-83.8(7)
N5	C44	C52	O7	53.2(3)	C44	C45	C46A	C51A	96.1(6)
N5	C44	C52	N6	-129.1(2)	C45	C44	C52	O7	-64.6(3)
N6	C53	C54	C55	-176.3(3)	C45	C44	C52	N6	113.1(2)
N6	C53	C58	C57	176.6(2)	C45	C46	C47	C48	179.9(5)
N6	C53	C58	C59	50.0(3)	C45	C46	C51	C50	-179.2(10)
C1	O1	C5	O2	2.7(4)	C45	C46A	C47A	C48A	179.0(12)
C1	O1	C5	N1	-176.3(2)	C45	C46A	C51A	C50A	-178.3(6)
C5	O1	C1	C2	-60.8(4)	C46	C45	C46A	C47A	11.0(7)
C5	O1	C1	C3	63.7(3)	C46	C45	C46A	C51A	-169.1(15)
C5	O1	C1	C4	-178.6(2)	C46	C47	C48	C49	-0.8(9)
C5	N1	C6	C7	-173.1(2)	C47	C46	C51	C50	1.3(12)
C5	N1	C6	C14	63.9(3)	C47	C48	C49	C50	1.5(12)
C6	N1	C5	O1	-172.1(2)	C48	C49	C50	C51	-0.8(19)
C6	N1	C5	O2	8.9(4)	C49	C50	C51	C46	-0.6(19)
C6	C7	C8	C9	-103.9(3)	C51	C46	C47	C48	-0.6(9)
C6	C7	C8	C13	77.3(3)	C46A	C45	C46	C47	166.8(14)
C7	C6	C14	O3	-86.6(3)	C46A	C45	C46	C51	-12.7(7)
C7	C6	C14	N2	93.3(2)	C46A	C47A	C48A	C49A	-1(2)
C7	C8	C9	C10	-177.1(3)	C47A	C46A	C51A	C50A	1.6(10)
C7	C8	C13	C12	177.2(2)	C47A	C48A	C49A	C50A	2(2)
C8	C9	C10	C11	-0.8(5)	C48A	C49A	C50A	C51A	-0.9(16)
C9	C8	C13	C12	-1.7(4)	C49A	C50A	C51A	C46A	-0.7(10)
C9	C10	C11	C12	-0.2(5)	C51A	C46A	C47A	C48A	-0.8(15)
C10	C11	C12	C13	0.3(5)	C52	N6	C53	C54	108.9(3)
C11	C12	C13	C8	0.7(5)	C52	N6	C53	C58	-127.4(2)
C13	C8	C9	C10	1.8(4)	C52	C44	C45	C46	-67.4(4)
C14	N2	C15	C16	95.6(3)	C52	C44	C45	C46A	-37.9(5)
C14	N2	C15	C20	-139.5(2)	C53	N6	C52	O7	5.3(4)
C14	C6	C7	C8	-163.99(19)	C53	N6	C52	C44	-172.3(2)
C15	N2	C14	O3	0.2(4)	C53	C54	C55	C56	-55.7(4)
C15	N2	C14	C6	-179.7(2)	C53	C58	C59	C60	53.1(3)
C15	C16	C17	C18	-56.4(3)	C53	C58	C59	C62	-178.7(2)
C15	C20	C21	C22	143.0(2)	C54	C53	C58	C57	-59.4(3)
C15	C20	C21	C24	-96.8(2)	C54	C53	C58	C59	174.0(2)
C16	C15	C20	C19	-58.2(3)	C54	C55	C56	C57	54.0(4)
C16	C15	C20	C21	175.0(2)	C55	C56	C57	C58	-55.7(4)
C16	C17	C18	C19	55.3(3)	C56	C57	C58	C53	57.8(3)
C17	C18	C19	C20	-56.9(3)	C56	C57	C58	C59	-176.2(2)
C18	C19	C20	C15	57.0(3)	C57	C58	C59	C60	-70.9(3)
C18	C19	C20	C21	-178.0(2)	C57	C58	C59	C62	57.3(3)
C19	C20	C21	C22	20.0(3)	C58	C53	C54	C55	58.8(3)
C19	C20	C21	C24	140.2(2)	C58	C59	C60	C61	-166.9(2)
C20	C15	C16	C17	59.1(3)	C58	C59	C62	O8	63.5(4)
C20	C21	C22	C23	-179.1(2)	C58	C59	C62	O9	-115.3(3)
C20	C21	C24	O4	-63.5(3)	C60	C59	C62	O8	-166.9(3)
C20	C21	C24	N3	116.6(2)	C60	C59	C62	O9	14.3(3)
C22	C21	C24	O4	61.8(3)	C62	O9	C63	C64	154.6(3)
C22	C21	C24	N3	-118.1(2)	C62	O9	C63	C64A	-178.7(3)
C24	N3	C25	C26	-177.2(2)	C62	O9	C63	C64B	-160.6(10)

C24	N3	C25	C33	60.8(3)	C62	C59	C60	C61	68.1(3)
C24	C21	C22	C23	59.7(3)	C63	O9	C62	O8	0.4(4)
C25	N3	C24	O4	8.7(4)	C63	O9	C62	C59	179.2(3)
C25	N3	C24	C21	-171.4(2)	C63	C64	C65	C66	-172.6(5)
C25	C26	C27	C28	108.4(6)	C63	C64	C69	C68	171.9(5)
C25	C26	C27	C32	-73.4(5)	C63	C64A	C65A	C66A	-173.9(5)
C25	C26	C27A	C28A	90.9(8)	C63	C64A	C69A	C68A	174.0(5)
C25	C26	C27A	C32A	-84.8(4)	C63	C64B	C65B	C66B	-178.3(19)
C26	C25	C33	O5	-87.7(3)	C63	C64B	C69B	C68B	178(3)
C26	C25	C33	N4	90.8(3)	C64	C63	C64A	C65A	156.3(8)
C26	C27	C28	C29	178.3(7)	C64	C63	C64A	C69A	-17.7(5)
C26	C27	C32	C31	-178.2(7)	C64	C63	C64B	C65B	158.1(19)
C26	C27A	C28A	C29A	-175.5(9)	C64	C63	C64B	C69B	-19.9(14)
C26	C27A	C32A	C31A	176.0(7)	C64	C65	C66	C67	0.0
C27	C26	C27A	C28A	-157(7)	C65	C64	C69	C68	0.0
C27	C26	C27A	C32A	28(7)	C65	C66	C67	C68	0.0
C27	C28	C29	C30	0.0	C66	C67	C68	C69	0.0
C28	C27	C32	C31	0.0	C67	C68	C69	C64	0.0
C28	C29	C30	C31	0.0	C69	C64	C65	C66	0.0
C29	C30	C31	C32	0.0	C64A	C63	C64	C65	94.4(7)
C30	C31	C32	C27	0.0	C64A	C63	C64	C69	-77.9(7)
C32	C27	C28	C29	0.0	C64A	C63	C64B	C65B	175(4)
C27A	C26	C27	C28	38(7)	C64A	C63	C64B	C69B	-3.3(18)
C27A	C26	C27	C32	-144(7)	C64A	C65A	C66A	C67A	0.0
C27A	C28A	C29A	C30A	0.0	C65A	C64A	C69A	C68A	0.0
C28A	C27A	C32A	C31A	0.0	C65A	C66A	C67A	C68A	0.0
C28A	C29A	C30A	C31A	0.0	C66A	C67A	C68A	C69A	0.0
C29A	C30A	C31A	C32A	0.0	C67A	C68A	C69A	C64A	0.0
C30A	C31A	C32A	C27A	0.0	C69A	C64A	C65A	C66A	0.0
C32A	C27A	C28A	C29A	0.0	C64B	C63	C64	C65	108.5(15)
C33	N4	C34	C35	121.1(2)	C64B	C63	C64	C69	-63.8(15)
C33	N4	C34	C39	-116.3(2)	C64B	C63	C64A	C65A	5(3)
C33	C25	C26	C27	-77.7(5)	C64B	C63	C64A	C69A	-169(3)
C33	C25	C26	C27A	-70.6(5)	C64B	C65B	C66B	C67B	0.0
C34	N4	C33	O5	-2.1(4)	C65B	C64B	C69B	C68B	0.0
C34	N4	C33	C25	179.4(2)	C65B	C66B	C67B	C68B	0.0
C34	C35	C36	C37	-58.0(3)	C66B	C67B	C68B	C69B	0.0
C34	C39	C40	C41	93.9(2)	C67B	C68B	C69B	C64B	0.0
C34	C39	C40	C43	-141.1(2)	C69B	C64B	C65B	C66B	0.0
C35	C34	C39	C38	-48.9(3)					

Table S65. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for gellman156_sq.

Atom	***x***	***y***	***z***	**U(eq)**
H1	7651	195	2665	34
H2	7410	-601	4288	32
H3	6719	1058	4017	35
H4	8131	2121	5399	33

H5	7077	982	5975	33
H6	6857	2101	7501	34
H2A	5712	572	2212	136
H2B	5343	1499	2036	136
H2C	5685	1345	2649	136
H3A	6433	2507	2830	116
H3B	6074	3018	2299	116
H3C	6781	2820	2386	116
H4A	6584	2041	1512	69
H4B	5868	2161	1414	69
H4C	6154	1160	1404	69
H6A	7730	285	3725	30
H7A	8378	-1081	3767	33
H7B	8262	-1011	3129	33
H9	8874	8	2747	46
H10	9696	1001	2863	60
H11	10109	1641	3700	62
H12	9695	1285	4425	53
H13	8876	293	4313	42
H15	6436	-1746	3901	33
H16A	7273	-2754	4155	41
H16B	7411	-2357	4754	41
H17A	6339	-3344	4246	52
H17B	6861	-3732	4726	52
H18A	6627	-2646	5320	55
H18B	6022	-3190	5048	55
H19A	5832	-1586	5054	45
H19B	5694	-1965	4452	45
H20	6789	-1049	4973	33
H21	6117	-164	4051	34
H22A	5286	-452	4455	44
H22B	5653	14	4999	44
H23A	5616	1443	4576	78
H23B	4942	1051	4467	78
H23C	5310	996	4008	78
H25	7217	2126	4852	35
H26A	7760	1897	3957	47
H26B	7143	2457	3887	47
H26C	7718	1884	3928	47
H26D	7146	2531	3912	47
H28	8650	2608	4203	54
H29	9250	3907	4466	62
H30	8831	5220	4759	65
H31	7813	5234	4790	63
H32	7213	3935	4528	52
H28A	8772	2514	4420	51
H29A	9328	3784	4820	61
H30A	8828	5101	5010	62
H31A	7771	5147	4800	63
H32A	7216	3877	4400	49
H34	8853	549	5549	34

H35A	9406	1780	5326	44
H35B	9217	2428	5763	44
H36A	10216	1911	6095	50
H36B	9998	864	6012	50
H37A	10114	1288	6926	47
H37B	9641	2111	6751	47
H38A	9142	845	7028	42
H38B	9354	197	6603	42
H39	8514	1679	6349	32
H40	7870	389	5851	32
H41A	8596	-808	6146	41
H41B	8518	-659	6745	41
H42A	7490	-1084	6477	71
H42B	7895	-1887	6319	71
H42C	7546	-1191	5870	71
H44	6719	972	6918	32
H45A	6107	1744	5894	40
H45B	6047	687	6039	40
H45C	6163	1455	5830	40
H45D	5950	677	6184	40
H47	5357	247	6579	51
H48	4537	568	6945	59
H49	4261	2077	7043	56
H50	4781	3268	6738	52
H51	5590	2937	6356	50
H47A	5347	1140	6837	49
H48A	4625	2167	7015	49
H49A	4506	3594	6613	56
H50A	5065	4003	5993	56
H51A	5771	2967	5799	48
H53	7020	3900	7199	40
H54A	6249	3417	7892	60
H54B	6012	3738	7282	60
H55A	6436	5203	7487	74
H55B	6006	4987	7884	74
H56A	6832	4631	8578	66
H56B	6947	5620	8349	66
H57A	7831	4687	8477	52
H57B	7604	4985	7862	52
H58	7343	3245	8280	41
H59	7990	2518	7855	44
H60A	7912	3523	7093	49
H60B	8400	4176	7467	49
H61A	9128	2998	7586	86
H61B	8867	3155	6957	86
H61C	8640	2302	7250	86
H63A	9516	3526	9075	69
H63B	9314	4584	9052	69
H63C	9650	3503	8982	69
H63D	9262	4344	9128	69
H63E	9751	3592	8918	69

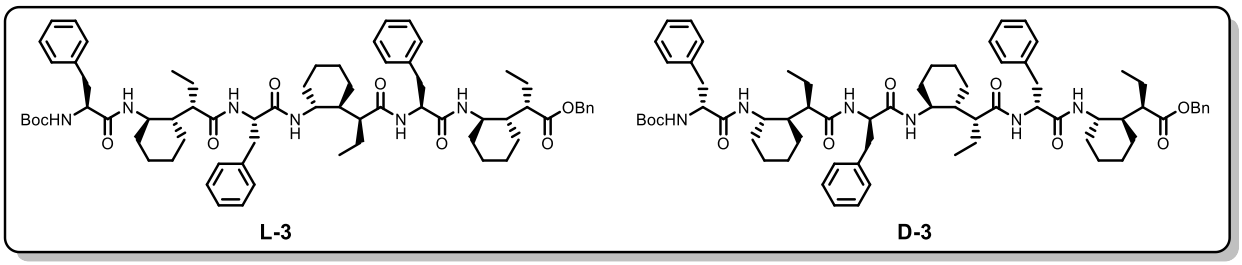
H63F	9278	4186	9165	69
H65	10518	4243	9427	56
H66	11402	4819	9233	71
H67	11408	5259	8357	85
H68	10529	5123	7675	81
H69	9644	4547	7869	63
H65A	9289	5818	8506	75
H66A	9945	6793	8190	114
H67A	10923	6301	8188	101
H68A	11244	4834	8501	94
H69A	10587	3859	8817	76
H65B	8758	5241	8453	75
H66B	8836	6659	8047	75
H67B	9783	7209	7984	75
H68B	10651	6341	8328	75
H69B	10574	4923	8734	75
H1S	8592	2897	9699	49
H1SA	7481	2916	5949	111
H1SB	7792	3691	5811	111

Table S66. Atomic Occupancy for gellman156_sq.

Atom	***Occupancy***	**Atom**	***Occupancy***	**Atom**	***Occupancy***
H26A	0.516(15)	H26B	0.516(15)	H26C	0.484(15)
H26D	0.484(15)	C27	0.516(15)	C28	0.516(15)
H28	0.516(15)	C29	0.516(15)	H29	0.516(15)
C30	0.516(15)	H30	0.516(15)	C31	0.516(15)
H31	0.516(15)	C32	0.516(15)	H32	0.516(15)
C27A	0.484(15)	C28A	0.484(15)	H28A	0.484(15)
C29A	0.484(15)	H29A	0.484(15)	C30A	0.484(15)
H30A	0.484(15)	C31A	0.484(15)	H31A	0.484(15)
C32A	0.484(15)	H32A	0.484(15)	H45A	0.543(5)
H45B	0.543(5)	H45C	0.457(5)	H45D	0.457(5)
C46	0.543(5)	C47	0.543(5)	H47	0.543(5)
C48	0.543(5)	H48	0.543(5)	C49	0.543(5)
H49	0.543(5)	C50	0.543(5)	H50	0.543(5)
C51	0.543(5)	H51	0.543(5)	C46A	0.457(5)
C47A	0.457(5)	H47A	0.457(5)	C48A	0.457(5)
H48A	0.457(5)	C49A	0.457(5)	H49A	0.457(5)
C50A	0.457(5)	H50A	0.457(5)	C51A	0.457(5)
H51A	0.457(5)	H63A	0.450(2)	H63B	0.450(2)
H63C	0.442(3)	H63D	0.442(3)	H63E	0.109(3)
H63F	0.109(3)	C64	0.450(2)	C65	0.450(2)
H65	0.450(2)	C66	0.450(2)	H66	0.450(2)
C67	0.450(2)	H67	0.450(2)	C68	0.450(2)
H68	0.450(2)	C69	0.450(2)	H69	0.450(2)
C64A	0.442(3)	C65A	0.442(3)	H65A	0.442(3)
C66A	0.442(3)	H66A	0.442(3)	C67A	0.442(3)
H67A	0.442(3)	C68A	0.442(3)	H68A	0.442(3)

C69A	0.442(3)	H69A	0.442(3)	C64B	0.109(3)
C65B	0.109(3)	H65B	0.109(3)	C66B	0.109(3)
H66B	0.109(3)	C67B	0.109(3)	H67B	0.109(3)
C68B	0.109(3)	H68B	0.109(3)	C69B	0.109(3)
H69B	0.109(3)				

Crystallographic Experimental Section for Compound *rac*-3b



Data Collection

A colorless crystal with approximate dimensions 0.790 x 0.548 x 0.358 mm^3 was selected from the mother liquor at -15° C using a low temperature basin filled with dry-ice under a constant flow of argon to provide an inert atmosphere. The crystal was attached to the tip of a MiTeGen MicroMesh©. The crystal was mounted in a stream of cold nitrogen at 100(1) K and centered in the X-ray beam by using a video camera.

The crystal evaluation and data collection were performed on a Bruker SMART APEXII diffractometer with Cu K_α (λ = 1.54178 Å) radiation and the diffractometer to crystal distance of 4.03 cm.

The initial cell constants were obtained from three series of ω scans at different starting angles. Each series consisted of 35 frames collected at intervals of 0.7º in a 25º range about ω with the exposure time of 3 seconds per frame. The reflections were successfully indexed by an automated indexing routine built in the APEXII program. The final cell constants were calculated from a set of 9122 strong reflections from the actual data collection.

The data were collected by using the full sphere data collection routine to survey the reciprocal space to the extent of a full sphere to a resolution of 0.82 Å. A total of 137980 data were harvested by collecting 19 sets of frames with 0.7º scans in ω and φ with an exposure time 8-15 sec per frame. These highly redundant datasets were corrected for Lorentz and polarization effects. The absorption correction was based on fitting a function to the empirical transmission surface as sampled by multiple equivalent measurements. [1]

Structure Solution and Refinement

The systematic absences in the diffraction data were uniquely consistent for the space group $P2_1/c$ that yielded chemically reasonable and computationally stable results of refinement [2-4].

A successful solution by the direct methods provided most non-hydrogen atoms from the *E*-map. The remaining non-hydrogen atoms were located in an alternating series of least-squares cycles and difference Fourier maps. All non-hydrogen atoms except where noted were refined with anisotropic displacement coefficients. All hydrogen atoms were included in the structure factor calculation at idealized

positions and were allowed to ride on the neighboring atoms with relative isotropic displacement coefficients.

The asymmetric unit consists of one molecule of the foldamer shown in Figure 1, 1.6 molecules of chloroform, and 1.4 molecules of pentane. The foldamer has positional disorder in the benzyl group C63 (major component: 89.3(10)%). Bond distance restraints and constraints as well as thermal parameter restraints were used to ensure a chemically reasonable and computationally stable refinement.

There are two compositionally and positionally disordered solvent molecule locations in the asymmetric unit, contributing 0.6 molecules of $CHCl_3$ and 1.4 molecules pentane to the overall asymmetric unit composition. Disordered solvent position C71 contains two positions of pentane (occupancies: 68.7(3))%, 15.5(3)%) and one positions of chloroform (occupancy: 15.82(19)%). Disordered solvent position C76 contains one position of pentane (occupancy: 55.9(2)%) and three positions of chloroform (occupancies 22.13(17)%, 14.63(17)%, 7.38(17)%). Disordered chloroform molecule C76C was refined isotropically. All disordered solvent molecules were modelled with idealized geometries as well as thermal parameter restraints and constraints [5].

The foldamer's conformation is stabilized by the presence of discrete hydrogen bonds between N-H and O atoms of alternating amide groups (e.g. the amide group N1 forms a hydrogen bond with amide group N3). This pattern is broken by amide groups N4 and N6, which are not oriented to form a hydrogen bond. This intramolecular hydrogen bonding motif is propagated in the intermolecular interactions. The penultimate amide group forms a hydrogen bond with the first amide group, and the last amide group forms a hydrogen bond with the second amide group of an adjacent molecule (symmetry code: x, 3/2-y, z+1/2).

The final least-squares refinement of 1076 parameters against 15878 data resulted in residuals *R* (based on F^2 for $I \geq 2\sigma$) and *wR* (based on F^2 for all data) of 0.0573 and 0.1488, respectively. The final difference Fourier map contained several peaks of electron density not exceeding ca. 1.2 e^-/A^3 in the vicinity of the disordered solvent molecules and considered noise.

Summary

Crystal Data for $C_{69}H_{94}N_6O_9$, 1.6($CHCl_3$), 1.4(C_5H_{12}) (*M* =1443.83): monoclinic, space group $P2_1/c$ (no. 14), *a* = 11.9161(8) Å, *b* = 27.097(4) Å, *c* = 25.5162(16) Å, β = 97.111(4)°, *V* = 8175.4(14) $Å^3$, *Z* = 4, *T* = 100.0 K, μ(CuKα) = 2.001 mm^{-1}, *Dcalc* = 1.173 g/mm^3, 137980 reflections measured ($6.524 \leq 2\Theta \leq 146.448$), 15878 unique ($R_{int}$ = 0.0239, R_{sigma} = 0.0116) which were used in all calculations. The final R_1 was 0.0573 ($I > 2\sigma(I)$) and wR_2 was 0.1488 (all data).

References

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[4] Guzei, I.A. (**2013**). Internal laboratory computer programs Gn.

[5] Guzei, I. A. (**2014**). *J. Appl. Cryst.* **47**, 806-809.

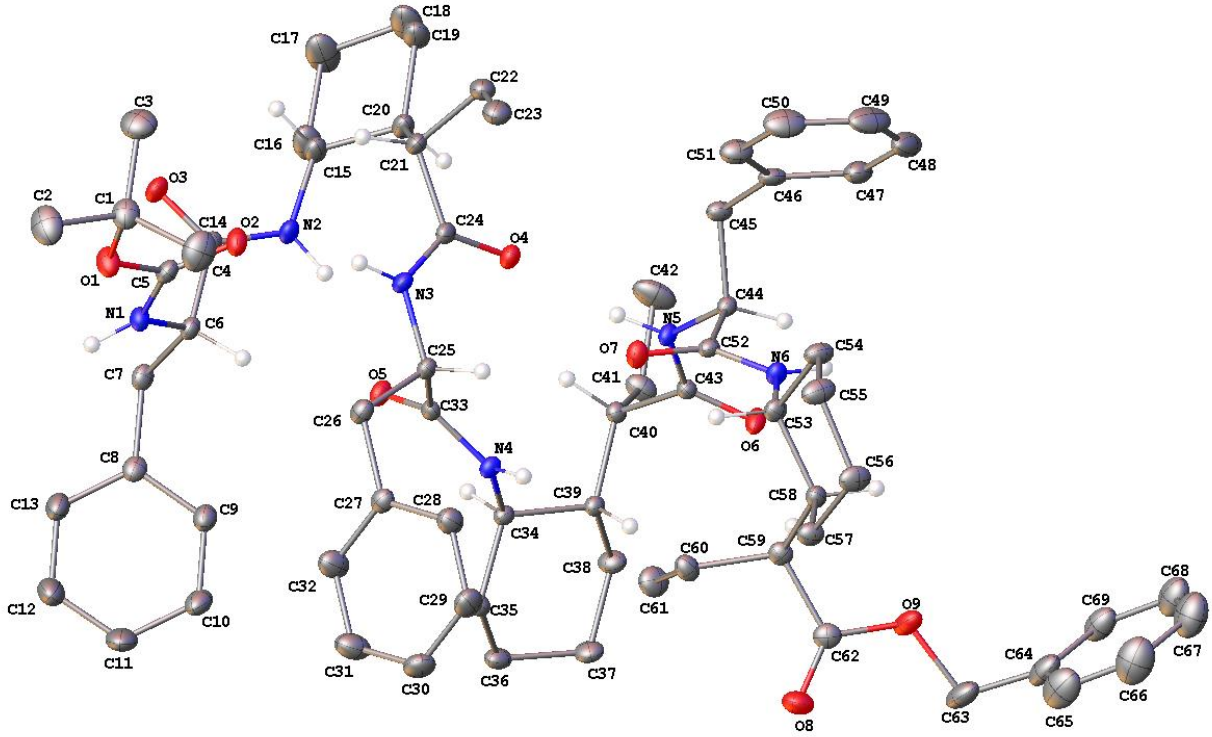

Figure S37. The foldamer of Gellman157. All atoms are drawn as 40% thermal probability ellipsoids. All disordered components and H atoms are omitted except those on heteroatoms and chiral carbons are omitted for clarity.

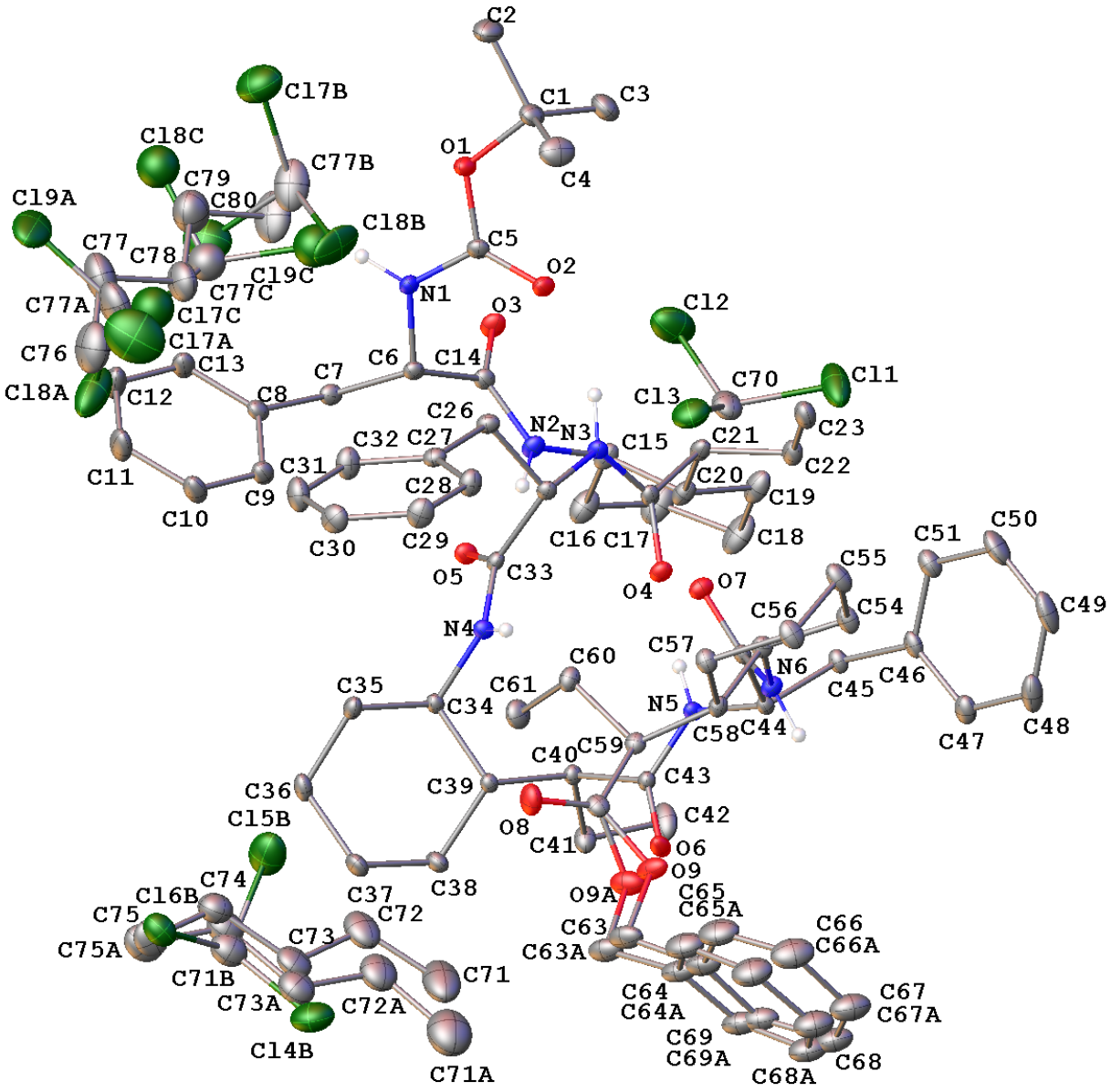

Figure S38. The asymmetric unit of Gellman157 showing all disordered components and solvent molecules. All atoms are drawn as 30% thermal probability ellipsoids. All H atoms except those on heteroatoms are omitted for clarity.

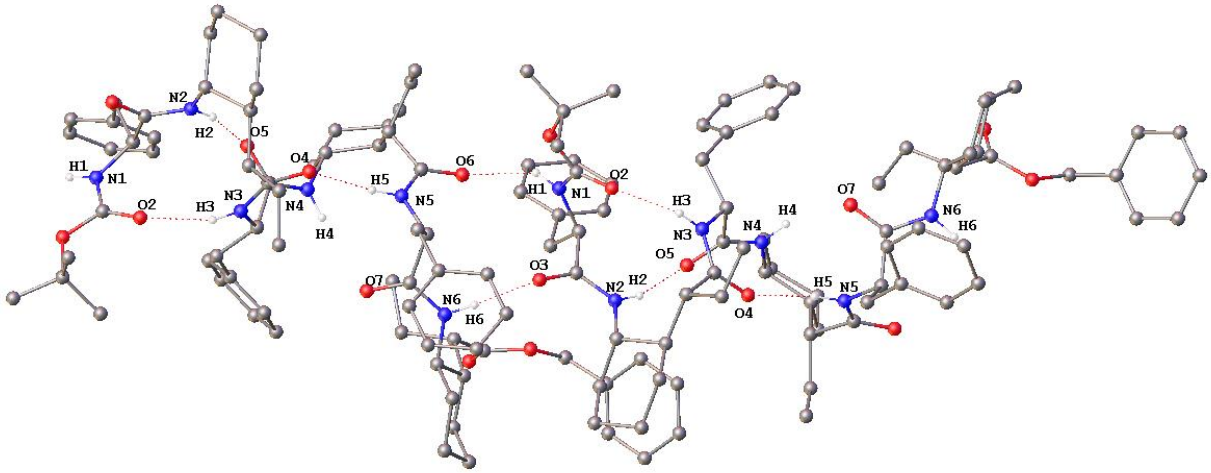

Figure S39. The intermolecular packing diagram for Gellman157. All H atoms except those on heteroatoms are omitted for clarity.

Table S67. Crystal data and structure refinement for Gellman157.	
Identification code	Gellman157
Empirical formula	$C_{69}H_{94}N_6O_9$, 1.6($CHCl_3$), 1.4(C_5H_{12})
Formula weight	1443.83
Temperature/K	100.0
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	11.9161(8)
b/Å	27.097(4)
c/Å	25.5162(16)
α/°	90
β/°	97.111(4)
γ/°	90
Volume/Å^3	8175.4(14)
Z	4
ρ_{calc}mg/mm^3	1.173
m/mm^{-1}	2.001
F(000)	3095.0
Crystal size/mm^3	0.79 × 0.548 × 0.358
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection	6.524 to 146.448°
Index ranges	$-14 \le h \le 14$, $-26 \le k \le 31$, $-31 \le l \le 30$
Reflections collected	137980
Independent reflections	15878 [R_{int} = 0.0239, R_{sigma} = 0.0116]
Data/restraints/parameters	15878/559/1076
Goodness-of-fit on F^2	1.062
Final R indexes [I>=2σ (I)]	R_1 = 0.0573, wR_2 = 0.1475
Final R indexes [all data]	R_1 = 0.0590, wR_2 = 0.1488
Largest diff. peak/hole / e Å^{-3}	1.13/-0.90

Table S68. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for Gellman157. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	*x*	*y*	*z*	U(eq)
O1	2097.3(11)	6457.4(5)	632.9(5)	26.7(3)
O2	1828.8(11)	6841.6(5)	1401.2(5)	24.3(3)
O3	2027.6(11)	7948.4(5)	849.5(5)	26.4(3)
O4	2066.2(11)	7658.0(5)	3128.8(5)	21.9(3)
O5	4239.0(11)	7636.2(5)	2560.5(5)	21.5(3)
O6	4015.9(11)	7821.8(6)	4921.2(5)	26.5(3)
O7	2407.2(11)	6540.0(5)	4047.6(5)	22.6(3)
O8	6464.4(12)	5758.7(6)	5602.6(6)	36.2(3)
N1	3265.0(12)	7056.5(6)	938.5(6)	19.6(3)
N2	2410.6(13)	8022.9(6)	1736.3(6)	21.2(3)
N3	2246.9(12)	7095.1(6)	2496.3(5)	18.3(3)
N4	4818.3(12)	7281.1(5)	3350.7(5)	17.1(3)
N5	2785.5(12)	7559.7(5)	4229.6(5)	16.7(3)

N6	2351.2(12)	6470.7(6)	4935.9(6)	18.7(3)
C1	1140.6(17)	6113.8(8)	653.4(8)	28.9(4)
C2	1228(2)	5785.3(10)	177.6(10)	43.2(6)
C3	45.3(18)	6401.5(9)	589.5(10)	39.7(5)
C4	1308(2)	5809.3(9)	1157.8(10)	44.0(6)
C5	2350.6(15)	6790.8(7)	1021.7(7)	20.9(4)
C6	3590.3(14)	7478.0(7)	1275.4(6)	18.4(3)
C7	4573.9(15)	7753.4(7)	1071.5(7)	21.4(4)
C8	5704.7(15)	7499.3(7)	1169.9(7)	22.5(4)
C9	6182.0(16)	7389.5(8)	1682.2(7)	27.3(4)
C10	7236.5(17)	7171.6(9)	1777.8(8)	34.6(5)
C11	7845.1(18)	7065.3(11)	1364.5(9)	44.2(6)
C12	7382.6(19)	7173.2(12)	851.9(9)	46.7(6)
C13	6315.9(17)	7386.6(9)	756.1(8)	33.4(5)
C14	2598.1(15)	7837.7(7)	1270.5(7)	19.9(3)
C15	1429.4(17)	8342.8(8)	1770.2(7)	26.6(4)
C16	1722(2)	8886.7(9)	1721.2(9)	38.8(5)
C17	643(3)	9197(1)	1673.3(11)	51.3(7)
C18	-18(2)	9098(1)	2138.2(10)	49.8(7)
C19	-251.5(19)	8549.4(9)	2203.0(9)	36.8(5)
C20	847.4(16)	8245.0(7)	2267.4(7)	25.6(4)
C21	678.9(15)	7687.0(7)	2363.2(7)	22.5(4)
C22	-310.8(16)	7564.0(8)	2676.4(8)	28.4(4)
C23	-401.9(16)	7015.5(9)	2792.0(8)	31.8(4)
C24	1734.3(14)	7479.5(7)	2689.8(6)	18.6(3)
C25	3277.2(14)	6895.3(6)	2783.9(6)	17.4(3)
C26	3725.5(15)	6484.7(7)	2446.5(7)	21.3(4)
C27	4764.0(15)	6226.6(7)	2716.3(7)	20.8(4)
C28	4711.4(17)	5936.1(8)	3160.1(8)	27.9(4)
C29	5668.5(19)	5699.8(8)	3402.4(8)	33.5(5)
C30	6692.2(18)	5751.1(8)	3208.4(9)	33.8(5)
C31	6756.9(18)	6041.7(9)	2771.5(10)	39.0(5)
C32	5795.7(18)	6274.7(8)	2523.5(9)	33.3(5)
C33	4152.0(14)	7309.3(6)	2889.7(6)	16.5(3)
C34	5718.4(14)	7635.0(7)	3524.3(6)	17.1(3)
C35	6875.3(15)	7391.0(7)	3516.7(7)	22.8(4)
C36	7832.3(15)	7738.1(8)	3730.0(8)	26.7(4)
C37	7673.5(15)	7907.3(8)	4284.2(8)	27.4(4)
C38	6542.3(15)	8172.3(7)	4282.3(8)	24.6(4)
C39	5548.7(14)	7838.0(6)	4072.1(6)	16.1(3)
C40	4354.2(14)	8080.6(6)	4056.2(6)	17.1(3)
C41	4345.9(16)	8634.3(7)	4180.0(8)	27.5(4)
C42	3151(2)	8843.9(9)	4111.0(12)	43.6(6)
C43	3695.6(14)	7809.5(6)	4443.0(6)	16.6(3)
C44	2065.0(13)	7275.1(6)	4538.7(6)	16.1(3)
C45	813.3(14)	7385.9(7)	4335.6(7)	20.6(4)
C46	-0.5(14)	7070.0(7)	4597.8(7)	22.5(4)
C47	-179.9(16)	7149.4(8)	5118.5(8)	30.2(4)
C48	-891.5(18)	6836.9(11)	5360.1(9)	42.3(6)
C49	-1433.1(18)	6448.4(10)	5083.0(11)	46.5(6)
C50	-1273.7(18)	6374.0(9)	4565.0(11)	41.8(6)

C51	-558.3(16)	6680.6(8)	4322.3(9)	29.8(4)
C52	2299.5(13)	6723.4(6)	4481.9(6)	16.7(3)
C53	2498.3(15)	5936.9(7)	4977.4(7)	19.6(3)
C54	1437.8(15)	5692.9(7)	5144.3(8)	26.4(4)
C55	1577.8(17)	5133.2(8)	5190.9(9)	30.8(4)
C56	2642.4(16)	4994.6(7)	5557.5(8)	27.5(4)
C57	3688.5(15)	5244.1(7)	5383.6(7)	22.4(4)
C58	3542.0(14)	5804.4(7)	5369.3(7)	18.5(3)
C59	4616.6(15)	6082.1(6)	5253.6(7)	19.5(3)
C60	4959.6(16)	6014.1(7)	4700.3(7)	23.5(4)
C61	5543.5(18)	6475.0(8)	4527.8(9)	32.2(4)
C62	5601.7(15)	5959.1(7)	5675.0(7)	22.7(4)
O9	5350(2)	6093.0(13)	6153.7(7)	33.3(7)
C63	6211(3)	6022.2(16)	6608.4(12)	38.2(8)
C64	5598(3)	5885.4(10)	7067.9(10)	37.1(7)
C65	5473(4)	5390.8(9)	7193.1(9)	50.6(9)
C66	4844(4)	5259.0(7)	7595.3(9)	57.7(10)
C67	4341(4)	5621.9(8)	7872.4(8)	53.2(10)
C68	4465(3)	6116.5(8)	7747.2(11)	44.6(8)
C69	5094(3)	6248.3(8)	7344.9(12)	37.7(7)
O9A	5640(20)	6257(8)	6089(7)	38(3)
C63A	6460(20)	6167(14)	6558(8)	38(3)
C64A	5870(20)	5965(9)	7003(10)	38(3)
C69A	5330(30)	6307(7)	7290(13)	42(3)
C68A	4830(20)	6154(8)	7727(11)	46(3)
C67A	4870(30)	5661(8)	7877(8)	52(3)
C66A	5420(30)	5320(7)	7590(9)	53(3)
C65A	5920(30)	5472(9)	7154(9)	48(3)
Cl1	-100.1(6)	5592.2(3)	3561.5(5)	79.5(3)
Cl2	1338.5(8)	5641.5(4)	2734.5(3)	77.5(3)
Cl3	2095.4(6)	5137.7(2)	3705.0(3)	50.97(16)
C70	1300(2)	5635.8(9)	3418.2(11)	43.0(6)
C71	6444(5)	9686(2)	4156(3)	88.5(19)
C72	7099(4)	9521.9(17)	3720(2)	63.3(12)
C73	8258(4)	9328.2(18)	3905(2)	50.5(10)
C74	8905(5)	9133(2)	3473(2)	51.7(11)
C75	10018(7)	8915(4)	3664(4)	50(3)
C71A	6785(18)	9614(10)	4580(9)	97(6)
C72A	7210(14)	9540(7)	4043(8)	63(3)
C73A	8417(15)	9335(7)	4085(7)	55(3)
C74A	8852(18)	9260(8)	3550(8)	57(3)
C75A	10060(20)	9056(11)	3597(12)	59(5)
Cl4B	8432(5)	9359(2)	4308.4(15)	65.4(15)
Cl6B	9930(6)	8787(4)	3710(3)	48(2)
Cl5B	7836(4)	9210(2)	3170.4(15)	74.5(17)
C71B	8957(3)	9286(2)	3691.2(18)	57(3)
C76	8593(4)	5222(2)	1794(2)	79.6(19)
C77	7908(3)	5330.4(19)	1257.9(16)	76.5(17)
C78	6807(3)	5611.2(13)	1302.4(13)	55.3(12)
C79	6114(3)	5722.0(19)	768.6(14)	75.9(16)
C80	5017(3)	6003(2)	818(2)	74.5(19)

Cl7A	8224(6)	4885.7(14)	2027.3(16)	120(2)
Cl8A	8500(4)	5902.8(10)	1673.3(16)	90.0(16)
Cl9A	8509(4)	5084.6(14)	922.3(15)	88.9(16)
C77A	7958(3)	5307.8(11)	1493.2(13)	71(3)
Cl7B	4614(6)	5310(2)	310.5(17)	85(2)
Cl8B	4749(6)	5578(2)	1429.4(16)	76(2)
Cl9B	5840(6)	6200.9(19)	701(2)	86(2)
C77B	4684(4)	5802.9(15)	772.1(16)	72(6)
Cl9C	4901(9)	5740(6)	1284(5)	83(2)
Cl7C	7359(9)	5600(6)	1424(4)	83(2)
Cl8C	6150(10)	5751(5)	364(4)	83(2)
C77C	6196(8)	5894(3)	1046(4)	66(4)

Table S69. Anisotropic Displacement Parameters ($Å^2×10^3$) for Gellman157. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	**U_{11}**	**U_{22}**	**U_{33}**	**U_{23}**	**U_{13}**	**U_{12}**
O1	26.6(7)	32.7(8)	21.6(6)	-7.6(5)	5.4(5)	-9.8(6)
O2	25.1(6)	32.4(7)	15.8(6)	-1.8(5)	3.8(5)	-5.0(5)
O3	28.2(7)	34.5(8)	15.4(6)	4.8(5)	-1.4(5)	7.0(6)
O4	23.4(6)	27.6(7)	14.0(6)	0.4(5)	0.2(5)	3.6(5)
O5	24.2(6)	25.1(7)	14.8(6)	4.9(5)	0.6(5)	-2.5(5)
O6	19.9(6)	45.5(9)	13.8(6)	0.4(5)	1.2(5)	-8.0(6)
O7	29.8(7)	21.3(7)	17.0(6)	1.2(5)	3.9(5)	1.2(5)
O8	21.7(7)	49.1(10)	38.1(8)	4.7(7)	5.4(6)	9.0(6)
N1	20.2(7)	24.3(8)	14.6(7)	-1.5(6)	3.1(5)	-1.0(6)
N2	25.0(8)	24.7(8)	13.5(7)	3.8(6)	0.9(6)	5.8(6)
N3	17.2(7)	23.2(8)	13.6(6)	0.9(5)	-1.8(5)	1.3(6)
N4	18.2(7)	17.7(7)	14.9(7)	3.1(5)	-0.2(5)	-0.2(5)
N5	17.3(7)	20.4(8)	12.1(6)	2.8(5)	0.7(5)	-2.2(5)
N6	22.0(7)	18.6(8)	15.2(7)	2.4(5)	1.8(5)	4.0(6)
C1	29.5(10)	29.0(11)	28.2(10)	-2.2(8)	3.7(8)	-9.9(8)
C2	47.2(13)	42.4(14)	40.8(13)	-15(1)	8.7(10)	-17.1(11)
C3	27.7(11)	43.7(14)	46.4(13)	-4.2(10)	-0.7(9)	-8.4(9)
C4	60.5(16)	30.7(13)	40.1(13)	6(1)	4.0(11)	-8.3(11)
C5	21.8(8)	24.4(10)	15.3(8)	1.0(7)	-2.2(6)	0.5(7)
C6	20.1(8)	21.2(9)	13.4(7)	1.5(6)	0.1(6)	-0.4(7)
C7	25.7(9)	21.1(9)	17.4(8)	3.3(6)	2.7(7)	-3.2(7)
C8	20.5(8)	25.8(10)	21.1(9)	1.1(7)	1.6(7)	-7.3(7)
C9	23.5(9)	38.1(12)	20.3(9)	1.2(8)	2.7(7)	-4.5(8)
C10	21.7(9)	55.9(14)	25(1)	5.7(9)	-1.7(8)	-3.9(9)
C11	18.5(10)	77.1(19)	36.6(12)	2.5(12)	1.3(8)	6.9(10)
C12	25.8(11)	86(2)	28.9(11)	-2.0(12)	6.6(9)	6.5(11)
C13	24.3(10)	55.9(14)	19.7(9)	1.3(9)	1.5(7)	-1.9(9)
C14	21.3(8)	21.7(9)	16.2(8)	3.9(6)	0.7(6)	-0.1(7)
C15	31.2(10)	29.8(11)	18.6(9)	5.4(7)	3.0(7)	11.8(8)
C16	53.0(14)	30.7(12)	35.0(11)	10.3(9)	14.2(10)	14.2(10)
C17	70.1(18)	38.0(14)	49.2(15)	17.7(11)	21.1(13)	27.7(13)
C18	64.9(17)	43.0(15)	44.9(14)	11.7(11)	20.2(12)	33.0(13)

C19	37.6(12)	44.9(13)	28.8(10)	7.3(9)	7.2(9)	21.9(10)
C20	28.7(10)	30.6(11)	17.3(8)	2.3(7)	2.4(7)	11.4(8)
C21	19.7(8)	32(1)	15.1(8)	0.1(7)	-0.4(6)	6.8(7)
C22	18.5(9)	42.2(12)	24.5(9)	1.1(8)	2.1(7)	5.7(8)
C23	20.1(9)	44.6(13)	30.7(10)	3.1(9)	3.1(8)	-0.2(8)
C24	17.0(8)	24.9(9)	14.0(8)	4.7(6)	2.4(6)	0.3(6)
C25	18.1(8)	20.0(9)	13.6(7)	2.2(6)	-0.1(6)	1.8(6)
C26	24.7(9)	20.2(9)	18.1(8)	-0.4(7)	-1.0(7)	1.5(7)
C27	24.2(9)	18.3(9)	19.3(8)	-2.3(6)	0.3(7)	2.2(7)
C28	27.6(10)	30.4(11)	26.3(9)	6.1(8)	6.0(8)	4.3(8)
C29	36.3(11)	35.7(12)	28.2(10)	10.5(8)	3.0(8)	9.2(9)
C30	28.5(10)	36.2(12)	35.3(11)	3.9(9)	-1.4(8)	11.6(9)
C31	25.9(10)	43.2(13)	49.4(13)	10.1(10)	10.1(9)	6.3(9)
C32	31(1)	34.1(12)	36.1(11)	13.1(9)	9.1(8)	5.5(8)
C33	16.6(8)	19.0(9)	14.4(7)	-0.2(6)	3.3(6)	4.0(6)
C34	14.6(8)	21.1(9)	15.6(8)	2.6(6)	1.1(6)	-0.3(6)
C35	18.2(8)	31.4(10)	19.1(8)	-0.1(7)	3.5(7)	5.1(7)
C36	15.1(8)	37.6(11)	27.8(10)	8.0(8)	3.8(7)	1.5(7)
C37	16.2(8)	35.3(11)	29.2(10)	-1.4(8)	-3.1(7)	-1.9(7)
C38	18.5(8)	26.5(10)	27.8(9)	-3.8(7)	-1.5(7)	-2.3(7)
C39	15.0(8)	18.3(9)	15.0(8)	2.4(6)	1.9(6)	-0.7(6)
C40	16.3(8)	19.2(9)	16.2(8)	2.2(6)	3.2(6)	-0.3(6)
C41	25.6(9)	19.6(10)	38.9(11)	0.7(8)	10.1(8)	0.3(7)
C42	34.3(12)	25.1(12)	74.1(17)	2.7(11)	17.6(11)	8.4(9)
C43	15.1(8)	19.3(9)	15.5(8)	1.0(6)	2.8(6)	1.4(6)
C44	14.6(8)	19.3(9)	14.2(7)	2.2(6)	0.9(6)	-1.3(6)
C45	15.8(8)	20.9(9)	24.6(9)	5.8(7)	1.0(7)	1.5(6)
C46	12.6(8)	25(1)	29.4(9)	9.2(7)	0.6(7)	3.0(7)
C47	19.5(9)	41.6(12)	29.2(10)	8.2(8)	1.7(7)	4.7(8)
C48	24.2(10)	68.5(17)	35.4(12)	21.9(11)	8.8(9)	8.4(10)
C49	21(1)	54.9(16)	64.5(16)	31.1(13)	8.9(10)	-3.6(10)
C50	24.3(10)	36.2(13)	64.2(16)	11.9(11)	2.1(10)	-9.5(9)
C51	21.4(9)	28.4(11)	39.1(11)	5.8(8)	1.6(8)	-1.9(8)
C52	13.0(7)	20.1(9)	16.5(8)	2.1(6)	0.1(6)	0.1(6)
C53	21.5(8)	18.3(9)	18.7(8)	4.5(6)	2.0(6)	1.6(7)
C54	19.1(9)	29.2(11)	30(1)	10.2(8)	0.0(7)	1.5(7)
C55	23.6(9)	29.5(11)	38.0(11)	12.4(8)	-1.8(8)	-5.6(8)
C56	25.4(9)	21.5(10)	35(1)	12.9(8)	1.8(8)	-1.0(7)
C57	21.3(9)	19.4(9)	26.4(9)	5.6(7)	2.5(7)	1.7(7)
C58	19.6(8)	20.2(9)	16.0(8)	2.8(6)	3.5(6)	2.6(6)
C59	21.8(8)	14.6(8)	22.3(8)	1.4(6)	3.4(7)	1.7(6)
C60	26.2(9)	20.6(10)	25.2(9)	3.6(7)	8.8(7)	2.3(7)
C61	31.3(10)	32.2(12)	34.3(11)	8.3(8)	8.8(8)	-4.7(8)
C62	23.7(9)	15.6(9)	28.6(9)	0.8(7)	1.9(7)	-1.2(7)
O9	34.9(12)	37.3(15)	24.8(9)	-8.1(8)	-8.4(7)	17.6(10)
C63	38.6(16)	40(2)	31.6(13)	-7.7(11)	-14.0(11)	14.2(13)
C64	46.9(18)	33.6(16)	26.5(13)	-6.2(10)	-13.1(11)	9.8(12)
C65	70(2)	31.6(16)	47.0(16)	-12.4(12)	-4.5(16)	8.5(15)
C66	87(3)	32.7(16)	51.2(17)	-3.8(12)	1.8(18)	-4.0(16)
C67	77(3)	41.9(16)	38.7(14)	-6.3(12)	1.0(15)	-5.8(16)
C68	63(2)	37.3(15)	31.2(13)	-8.1(11)	-2.0(14)	3.1(14)

C69	51.4(19)	27.3(13)	30.8(15)	-5.2(10)	-8.8(13)	7.4(12)
O9A	41(6)	34(6)	35(5)	-5(5)	-13(5)	11(5)
C63A	42(5)	38(6)	31(5)	-7(5)	-13(5)	13(5)
C64A	47(5)	32(4)	30(4)	-9(4)	-11(4)	13(4)
C69A	53(5)	36(5)	33(5)	-9(5)	-10(5)	8(5)
C68A	64(5)	35(5)	36(5)	-9(4)	-3(5)	2(5)
C67A	74(5)	38(5)	42(5)	-8(4)	0(5)	-2(5)
C66A	75(6)	34(5)	49(5)	-10(5)	1(5)	3(5)
C65A	64(6)	34(5)	42(5)	-11(5)	-5(5)	9(5)
Cl1	48.2(4)	46.9(4)	150.3(9)	-34.8(5)	39.2(5)	-11.6(3)
Cl2	84.3(6)	94.8(7)	50.3(4)	12.3(4)	-4.3(4)	-29.4(5)
Cl3	67.1(4)	35.5(3)	49.8(3)	-9.5(2)	5.6(3)	4.8(3)
C70	40.0(13)	32.3(13)	56.9(15)	-6.4(10)	6.5(11)	-8.6(10)
C71	70(3)	53(3)	146(6)	-39(3)	32(4)	-18(2)
C72	52(2)	38(2)	99(4)	-8(2)	8(2)	-11.4(17)
C73	48(2)	39.7(19)	63(3)	-1.7(19)	2.8(19)	-18.5(16)
C74	52(2)	51(3)	51(2)	-3.6(19)	0.9(18)	-9(2)
C75	54(4)	57(6)	39(3)	23(3)	3(2)	-2(3)
C71A	94(12)	69(11)	131(13)	-10(12)	23(12)	-15(10)
C72A	62(5)	43(5)	83(6)	-3(5)	12(5)	-16(5)
C73A	56(5)	43(5)	66(6)	0(6)	4(6)	-11(5)
C74A	56(5)	52(5)	59(5)	2(5)	0(5)	-9(5)
C75A	64(8)	59(9)	53(8)	18(8)	-1(7)	-8(8)
Cl4B	82(3)	54(3)	57(3)	-15(2)	-2(3)	1(2)
Cl6B	43(3)	49(4)	50(3)	9(3)	-1(2)	-6(2)
Cl5B	57(3)	108(4)	56(3)	13(3)	-2(2)	23(3)
C71B	58(5)	50(5)	61(5)	3(5)	2(5)	-9(5)
C76	64(4)	80(4)	99(5)	19(4)	23(3)	31(3)
C77	44(3)	76(4)	111(5)	19(3)	16(3)	1(3)
C78	36(2)	58(3)	72(3)	6(2)	6(2)	15(2)
C79	68(3)	89(4)	72(3)	8(3)	17(3)	-2(3)
C80	70(4)	47(4)	111(5)	23(3)	28(3)	31(3)
Cl7A	136(5)	89(4)	127(5)	43(3)	-20(4)	5(3)
Cl8A	122(4)	52(2)	112(3)	-11(2)	77(3)	-13(2)
Cl9A	91(3)	69(3)	114(3)	-38(2)	42(3)	-50(2)
C77A	36(5)	85(6)	90(7)	17(6)	1(5)	7(5)
Cl7B	98(5)	82(4)	75(4)	-18(3)	6(3)	12(4)
Cl8B	140(5)	58(3)	40(2)	7(2)	50(3)	-28(3)
Cl9B	97(5)	65(4)	93(4)	1(3)	-2(4)	5(3)
C77B	74(11)	44(11)	105(11)	30(10)	34(10)	23(9)

Table S70. Bond Lengths for Gellman157.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
O1	C1	1.478(2)	C44	C52	1.531(2)
O1	C5	1.348(2)	C45	C46	1.510(2)
O2	C5	1.222(2)	C46	C47	1.388(3)
O3	C14	1.235(2)	C46	C51	1.390(3)
O4	C24	1.239(2)	C47	C48	1.395(3)

O5	C33	1.234(2)	C48	C49	1.383(4)
O6	C43	1.233(2)	C49	C50	1.373(4)
O7	C52	1.236(2)	C50	C51	1.390(3)
O8	C62	1.197(2)	C53	C54	1.533(2)
N1	C5	1.345(2)	C53	C58	1.539(2)
N1	C6	1.453(2)	C54	C55	1.529(3)
N2	C14	1.334(2)	C55	C56	1.527(3)
N2	C15	1.467(2)	C56	C57	1.531(3)
N3	C24	1.333(2)	C57	C58	1.528(3)
N3	C25	1.455(2)	C58	C59	1.545(2)
N4	C33	1.337(2)	C59	C60	1.529(2)
N4	C34	1.466(2)	C59	C62	1.527(2)
N5	C43	1.336(2)	C60	C61	1.521(3)
N5	C44	1.456(2)	C62	O9	1.343(3)
N6	C52	1.340(2)	C62	O9A	1.326(9)
N6	C53	1.459(2)	O9	C63	1.464(3)
C1	C2	1.519(3)	C63	C64	1.503(4)
C1	C3	1.512(3)	C64	C65	1.3900
C1	C4	1.521(3)	C64	C69	1.3900
C6	C7	1.534(2)	C65	C66	1.3900
C6	C14	1.531(2)	C66	C67	1.3900
C7	C8	1.506(3)	C67	C68	1.3900
C8	C9	1.392(3)	C68	C69	1.3900
C8	C13	1.389(3)	O9A	C63A	1.464(10)
C9	C10	1.382(3)	C63A	C64A	1.504(10)
C10	C11	1.382(3)	C64A	C69A	1.3900
C11	C12	1.386(3)	C64A	C65A	1.3900
C12	C13	1.390(3)	C69A	C68A	1.3900
C15	C16	1.523(3)	C68A	C67A	1.3900
C15	C20	1.542(3)	C67A	C66A	1.3900
C16	C17	1.529(3)	C66A	C65A	1.3900
C17	C18	1.527(4)	Cl1	C70	1.756(3)
C18	C19	1.525(4)	Cl2	C70	1.751(3)
C19	C20	1.539(3)	Cl3	C70	1.756(3)
C20	C21	1.549(3)	C71	C72	1.503(8)
C21	C22	1.541(3)	C72	C73	1.498(6)
C21	C24	1.527(2)	C73	C74	1.514(7)
C22	C23	1.522(3)	C74	C75	1.479(8)
C25	C26	1.542(2)	C71A	C72A	1.5306
C25	C33	1.532(2)	C72A	C73A	1.5336
C26	C27	1.511(2)	C73A	C74A	1.5328
C27	C28	1.387(3)	C74A	C75A	1.5312
C27	C32	1.386(3)	Cl4B	C71B	1.7760
C28	C29	1.385(3)	Cl6B	C71B	1.7765
C29	C30	1.379(3)	Cl5B	C71B	1.7756
C30	C31	1.375(3)	C76	C77	1.5326
C31	C32	1.390(3)	C77	C78	1.5327
C34	C35	1.531(2)	C78	C79	1.5329
C34	C39	1.539(2)	C79	C80	1.5308
C35	C36	1.526(3)	Cl7A	C77A	1.7773
C36	C37	1.521(3)	Cl8A	C77A	1.7758

C37	C38	1.527(3)	Cl9A	C77A	1.7760
C38	C39	1.534(2)	Cl7B	C77B	1.7759
C39	C40	1.564(2)	Cl8B	C77B	1.7770
C40	C41	1.534(3)	Cl9B	C77B	1.7766
C40	C43	1.524(2)	Cl9C	C77C	1.7767
C41	C42	1.522(3)	Cl7C	C77C	1.7766
C44	C45	1.547(2)	Cl8C	C77C	1.7775

Table S71. Bond Angles for Gellman157.

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C5	O1	C1	119.98(14)	N5	C43	C40	115.94(14)
C5	N1	C6	119.32(14)	N5	C44	C45	108.94(13)
C14	N2	C15	119.84(15)	N5	C44	C52	109.83(13)
C24	N3	C25	120.04(14)	C52	C44	C45	109.74(14)
C33	N4	C34	124.10(14)	C46	C45	C44	112.77(14)
C43	N5	C44	123.43(14)	C47	C46	C45	121.28(18)
C52	N6	C53	124.15(15)	C47	C46	C51	118.74(18)
O1	C1	C2	102.20(16)	C51	C46	C45	119.95(18)
O1	C1	C3	109.32(17)	C46	C47	C48	120.2(2)
O1	C1	C4	110.54(17)	C49	C48	C47	120.5(2)
C2	C1	C4	110.1(2)	C50	C49	C48	119.5(2)
C3	C1	C2	111.08(19)	C49	C50	C51	120.4(2)
C3	C1	C4	113.07(19)	C50	C51	C46	120.6(2)
O2	C5	O1	124.70(17)	O7	C52	N6	124.81(17)
O2	C5	N1	124.51(17)	O7	C52	C44	121.19(15)
N1	C5	O1	110.78(15)	N6	C52	C44	113.99(14)
N1	C6	C7	109.94(14)	N6	C53	C54	110.61(15)
N1	C6	C14	110.26(14)	N6	C53	C58	111.02(14)
C14	C6	C7	108.02(14)	C54	C53	C58	110.29(14)
C8	C7	C6	115.24(15)	C55	C54	C53	111.33(16)
C9	C8	C7	120.49(17)	C56	C55	C54	111.47(17)
C13	C8	C7	121.08(17)	C55	C56	C57	111.00(15)
C13	C8	C9	118.38(18)	C58	C57	C56	110.54(15)
C10	C9	C8	120.93(18)	C53	C58	C59	112.62(14)
C11	C10	C9	120.37(19)	C57	C58	C53	109.16(15)
C10	C11	C12	119.4(2)	C57	C58	C59	113.17(14)
C11	C12	C13	120.1(2)	C60	C59	C58	116.14(15)
C8	C13	C12	120.78(19)	C62	C59	C58	110.01(14)
O3	C14	N2	123.27(17)	C62	C59	C60	110.72(15)
O3	C14	C6	120.21(16)	C61	C60	C59	110.64(16)
N2	C14	C6	116.51(15)	O8	C62	C59	126.18(18)
N2	C15	C16	112.01(17)	O8	C62	O9	123.75(19)
N2	C15	C20	112.95(15)	O8	C62	O9A	116.8(10)
C16	C15	C20	111.39(17)	O9	C62	C59	110.02(16)
C15	C16	C17	109.9(2)	O9A	C62	C59	112.2(9)
C18	C17	C16	110.56(19)	C62	O9	C63	118.2(2)
C19	C18	C17	112.0(2)	O9	C63	C64	106.8(2)
C18	C19	C20	111.7(2)	C65	C64	C63	119.56(19)

C15	C20	C21	112.21(15)	C65	C64	C69	120.0
C19	C20	C15	106.45(15)	C69	C64	C63	120.3(2)
C19	C20	C21	114.51(17)	C64	C65	C66	120.0
C22	C21	C20	114.55(16)	C67	C66	C65	120.0
C24	C21	C20	109.34(15)	C66	C67	C68	120.0
C24	C21	C22	105.59(14)	C69	C68	C67	120.0
C23	C22	C21	112.91(16)	C68	C69	C64	120.0
O4	C24	N3	122.03(16)	C62	O9A	C63A	120.4(18)
O4	C24	C21	119.92(16)	O9A	C63A	C64A	111(2)
N3	C24	C21	118.00(15)	C69A	C64A	C63A	116(2)
N3	C25	C26	108.41(13)	C69A	C64A	C65A	120.0
N3	C25	C33	109.25(14)	C65A	C64A	C63A	123(2)
C33	C25	C26	110.57(14)	C64A	C69A	C68A	120.0
C27	C26	C25	113.60(14)	C67A	C68A	C69A	120.0
C28	C27	C26	121.16(17)	C66A	C67A	C68A	120.0
C32	C27	C26	120.48(17)	C67A	C66A	C65A	120.0
C32	C27	C28	118.35(17)	C66A	C65A	C64A	120.0
C29	C28	C27	120.60(19)	Cl1	C70	Cl3	109.83(14)
C30	C29	C28	120.61(19)	Cl2	C70	Cl1	110.53(15)
C31	C30	C29	119.32(19)	Cl2	C70	Cl3	110.05(14)
C30	C31	C32	120.2(2)	C73	C72	C71	114.5(5)
C27	C32	C31	120.87(19)	C72	C73	C74	115.3(4)
O5	C33	N4	123.31(16)	C75	C74	C73	114.6(5)
O5	C33	C25	121.22(15)	C71A	C72A	C73A	113.3
N4	C33	C25	115.45(14)	C74A	C73A	C72A	113.8
N4	C34	C35	109.87(14)	C75A	C74A	C73A	113.2
N4	C34	C39	109.55(13)	Cl4B	C71B	Cl6B	111.2
C35	C34	C39	112.68(14)	Cl5B	C71B	Cl4B	111.2
C36	C35	C34	111.32(16)	Cl5B	C71B	Cl6B	111.2
C37	C36	C35	110.13(15)	C76	C77	C78	113.3
C36	C37	C38	110.52(15)	C77	C78	C79	113.8
C37	C38	C39	111.48(16)	C80	C79	C78	113.3
C34	C39	C40	110.39(13)	Cl8A	C77A	Cl7A	111.2
C38	C39	C34	110.26(14)	Cl8A	C77A	Cl9A	111.2
C38	C39	C40	114.92(14)	Cl9A	C77A	Cl7A	111.2
C41	C40	C39	115.78(14)	Cl7B	C77B	Cl8B	111.2
C43	C40	C39	108.71(13)	Cl7B	C77B	Cl9B	111.2
C43	C40	C41	108.74(14)	Cl9B	C77B	Cl8B	111.1
C42	C41	C40	111.77(17)	Cl9C	C77C	Cl8C	111.2
O6	C43	N5	123.59(15)	Cl7C	C77C	Cl9C	111.1
O6	C43	C40	120.46(15)	Cl7C	C77C	Cl8C	111.2

Table S72. Torsion Angles for Gellman157.

A	**B**	**C**	**D**	**Angle/°**	**A**	**B**	**C**	**D**	**Angle/°**
O8	C62	O9	C63	4.6(4)	C35	C34	C39	C38	52.15(19)
O8	C62	O9A	C63A	-29(3)	C35	C34	C39	C40	-179.80(14)
N1	C6	C7	C8	-73.71(19)	C35	C36	C37	C38	-59.0(2)
N1	C6	C14	O3	-44.8(2)	C36	C37	C38	C39	58.7(2)

N1	C6	C14	N2	136.23(16)	C37	C38	C39	C34	-54.5(2)
N2	C15	C16	C17	171.31(18)	C37	C38	C39	C40	-179.98(15)
N2	C15	C20	C19	-171.72(18)	C38	C39	C40	C41	7.9(2)
N2	C15	C20	C21	-45.7(2)	C38	C39	C40	C43	-114.79(16)
N3	C25	C26	C27	-176.76(15)	C39	C34	C35	C36	-53.8(2)
N3	C25	C33	O5	-37.0(2)	C39	C40	C41	C42	175.37(17)
N3	C25	C33	N4	144.67(14)	C39	C40	C43	O6	64.9(2)
N4	C34	C35	C36	-176.22(14)	C39	C40	C43	N5	-114.09(16)
N4	C34	C39	C38	174.79(14)	C41	C40	C43	O6	-61.9(2)
N4	C34	C39	C40	-57.16(17)	C41	C40	C43	N5	119.06(17)
N5	C44	C45	C46	174.47(15)	C43	N5	C44	C45	135.17(17)
N5	C44	C52	O7	-43.7(2)	C43	N5	C44	C52	-104.61(18)
N5	C44	C52	N6	137.46(14)	C43	C40	C41	C42	-61.9(2)
N6	C53	C54	C55	-179.88(15)	C44	N5	C43	O6	0.3(3)
N6	C53	C58	C57	-177.81(14)	C44	N5	C43	C40	179.26(15)
N6	C53	C58	C59	-51.23(19)	C44	C45	C46	C47	72.6(2)
C1	O1	C5	O2	-1.2(3)	C44	C45	C46	C51	-105.21(19)
C1	O1	C5	N1	178.08(16)	C45	C44	C52	O7	76.04(19)
C5	O1	C1	C2	-175.28(18)	C45	C44	C52	N6	-102.81(16)
C5	O1	C1	C3	67.0(2)	C45	C46	C47	C48	-176.71(18)
C5	O1	C1	C4	-58.1(2)	C45	C46	C51	C50	177.33(18)
C5	N1	C6	C7	-174.13(15)	C46	C47	C48	C49	-0.6(3)
C5	N1	C6	C14	-55.1(2)	C47	C46	C51	C50	-0.6(3)
C6	N1	C5	O1	172.10(14)	C47	C48	C49	C50	-0.6(3)
C6	N1	C5	O2	-8.6(3)	C48	C49	C50	C51	1.1(4)
C6	C7	C8	C9	-59.3(2)	C49	C50	C51	C46	-0.6(3)
C6	C7	C8	C13	123.3(2)	C51	C46	C47	C48	1.2(3)
C7	C6	C14	O3	75.4(2)	C52	N6	C53	C54	-113.71(18)
C7	C6	C14	N2	-103.62(18)	C52	N6	C53	C58	123.51(17)
C7	C8	C9	C10	-177.77(19)	C52	C44	C45	C46	54.20(19)
C7	C8	C13	C12	176.9(2)	C53	N6	C52	O7	-2.6(3)
C8	C9	C10	C11	1.0(4)	C53	N6	C52	C44	176.24(14)
C9	C8	C13	C12	-0.5(3)	C53	C54	C55	C56	54.1(2)
C9	C10	C11	C12	-0.9(4)	C53	C58	C59	C60	-56.7(2)
C10	C11	C12	C13	0.0(4)	C53	C58	C59	C62	176.52(15)
C11	C12	C13	C8	0.7(4)	C54	C53	C58	C57	59.22(19)
C13	C8	C9	C10	-0.3(3)	C54	C53	C58	C59	-174.20(15)
C14	N2	C15	C16	-92.8(2)	C54	C55	C56	C57	-54.3(2)
C14	N2	C15	C20	140.47(18)	C55	C56	C57	C58	57.6(2)
C14	C6	C7	C8	165.94(15)	C56	C57	C58	C53	-59.77(19)
C15	N2	C14	O3	5.3(3)	C56	C57	C58	C59	173.96(15)
C15	N2	C14	C6	-175.75(16)	C57	C58	C59	C60	67.7(2)
C15	C16	C17	C18	55.5(3)	C57	C58	C59	C62	-59.08(19)
C15	C20	C21	C22	-154.14(16)	C58	C53	C54	C55	-56.7(2)
C15	C20	C21	C24	87.59(18)	C58	C59	C60	C61	148.94(16)
C16	C15	C20	C19	61.2(2)	C58	C59	C62	O8	117.9(2)
C16	C15	C20	C21	-172.81(16)	C58	C59	C62	O9	-59.9(3)
C16	C17	C18	C19	-53.8(3)	C58	C59	C62	O9A	-87.7(16)
C17	C18	C19	C20	56.5(3)	C59	C62	O9	C63	-177.6(2)
C18	C19	C20	C15	-58.3(2)	C59	C62	O9A	C63A	173.6(16)
C18	C19	C20	C21	177.09(17)	C60	C59	C62	O8	-11.8(3)

C19	C20	C21	C22	-32.7(2)	C60	C59	C62	O9	170.4(2)
C19	C20	C21	C24	-150.92(16)	C60	C59	C62	O9A	142.6(16)
C20	C15	C16	C17	-61.1(2)	C62	C59	C60	C61	-84.68(19)
C20	C21	C22	C23	-176.37(16)	C62	O9	C63	C64	-145.0(3)
C20	C21	C24	O4	57.7(2)	C62	O9A	C63A	C64A	-106(3)
C20	C21	C24	N3	-124.73(17)	O9	C62	O9A	C63A	83(3)
C22	C21	C24	O4	-66.0(2)	O9	C63	C64	C65	95.6(2)
C22	C21	C24	N3	111.54(18)	O9	C63	C64	C69	-80.2(2)
C24	N3	C25	C26	-175.50(15)	C63	C64	C65	C66	-175.8(2)
C24	N3	C25	C33	-54.93(19)	C63	C64	C69	C68	175.8(2)
C24	C21	C22	C23	-56.0(2)	C64	C65	C66	C67	0.0
C25	N3	C24	O4	-4.7(3)	C65	C64	C69	C68	0.0
C25	N3	C24	C21	177.85(15)	C65	C66	C67	C68	0.0
C25	C26	C27	C28	66.9(2)	C66	C67	C68	C69	0.0
C25	C26	C27	C32	-113.2(2)	C67	C68	C69	C64	0.0
C26	C25	C33	O5	82.22(19)	C69	C64	C65	C66	0.0
C26	C25	C33	N4	-96.10(17)	O9A	C62	O9	C63	-78(2)
C26	C27	C28	C29	179.74(19)	O9A	C63A	C64A	C69A	-80(3)
C26	C27	C32	C31	179.5(2)	O9A	C63A	C64A	C65A	104(3)
C27	C28	C29	C30	0.3(3)	C63A	C64A	C69A	C68A	-176(2)
C28	C27	C32	C31	-0.6(3)	C63A	C64A	C65A	C66A	176(2)
C28	C29	C30	C31	0.3(4)	C64A	C69A	C68A	C67A	0.0
C29	C30	C31	C32	-1.0(4)	C69A	C64A	C65A	C66A	0.0
C30	C31	C32	C27	1.2(4)	C69A	C68A	C67A	C66A	0.0
C32	C27	C28	C29	-0.1(3)	C68A	C67A	C66A	C65A	0.0
C33	N4	C34	C35	-109.53(18)	C67A	C66A	C65A	C64A	0.0
C33	N4	C34	C39	126.17(16)	C65A	C64A	C69A	C68A	0.0
C33	C25	C26	C27	63.49(19)	C71	C72	C73	C74	176.6(4)
C34	N4	C33	O5	0.8(3)	C72	C73	C74	C75	-176.4(6)
C34	N4	C33	C25	179.05(14)	C71A	C72A	C73A	C74A	-179.9
C34	C35	C36	C37	56.5(2)	C72A	C73A	C74A	C75A	-180.0
C34	C39	C40	C41	-117.53(16)	C76	C77	C78	C79	-180.0
C34	C39	C40	C43	119.76(15)	C77	C78	C79	C80	-179.9

Table S73. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for Gellman157.

Atom	***x***	***y***	***z***	**U(eq)**
H1	3662	6976	683	24
H2	2874	7953	2022	25
H3	1962	6962	2194	22
H4	4709	7035	3563	21
H5	2612	7568	3884	20
H6	2293	6637	5227	22
H2A	1153	5987	-144	65
H2B	623	5538	151	65
H2C	1964	5619	219	65
H3A	28	6623	892	60
H3B	-594	6172	572	60

H3C	-8	6596	264	60
H4A	2079	5677	1209	66
H4B	764	5536	1131	66
H4C	1188	6019	1459	66
H6A	3827	7362	1645	22
H7A	4386	7805	686	26
H7B	4642	8083	1240	26
H9	5777	7465	1970	33
H10	7545	7094	2130	41
H11	8574	6920	1431	53
H12	7795	7101	566	56
H13	6001	7456	403	40
H15	864	8261	1459	32
H16A	2115	8937	1406	47
H16B	2237	8992	2036	47
H17A	841	9551	1665	62
H17B	165	9116	1339	62
H18A	417	9224	2466	60
H18B	-746	9279	2083	60
H19A	-751	8430	1890	44
H19B	-651	8501	2517	44
H20	1348	8379	2578	31
H21	561	7513	2015	27
H22A	-210	7748	3014	34
H22B	-1026	7677	2473	34
H23A	-454	6830	2460	48
H23B	-1080	6955	2964	48
H23C	269	6909	3026	48
H25	3107	6754	3127	21
H26A	3120	6237	2358	26
H26B	3908	6630	2111	26
H28	4013	5899	3299	33
H29	5619	5501	3705	40
H30	7346	5587	3375	41
H31	7461	6083	2639	47
H32	5847	6469	2218	40
H34	5659	7917	3269	21
H35A	6914	7087	3734	27
H35B	6969	7296	3150	27
H36A	8567	7566	3737	32
H36B	7840	8028	3495	32
H37A	8297	8133	4419	33
H37B	7699	7618	4523	33
H38A	6535	8471	4059	30
H38B	6453	8278	4646	30
H39	5564	7549	4316	19
H40	3946	8032	3693	21
H41A	4694	8689	4548	33
H41B	4807	8811	3943	33
H42A	2806	8792	3746	65
H42B	3180	9198	4189	65

H42C	2700	8676	4353	65
H44	2224	7372	4919	19
H45A	656	7738	4401	25
H45B	686	7330	3949	25
H47	183	7417	5311	36
H48	-1005	6891	5718	51
H49	-1912	6234	5250	56
H50	-1655	6111	4371	50
H51	-449	6624	3965	36
H53	2617	5807	4622	23
H54A	1287	5829	5489	32
H54B	780	5770	4881	32
H55A	908	4990	5329	37
H55B	1621	4992	4836	37
H56A	2743	4632	5555	33
H56B	2556	5097	5923	33
H57A	4364	5157	5632	27
H57B	3808	5124	5028	27
H58	3379	5909	5728	22
H59	4457	6442	5292	23
H60A	5477	5728	4699	28
H60B	4279	5945	4447	28
H61A	5826	6413	4189	48
H61B	6178	6560	4795	48
H61C	5003	6749	4488	48
H63A	6649	6330	6686	46
H63B	6740	5756	6536	46
H65	5817	5143	7004	61
H66	4759	4921	7681	69
H67	3911	5532	8147	64
H68	4121	6365	7937	54
H69	5179	6586	7259	45
H63C	6844	6479	6671	46
H63D	7034	5928	6471	46
H69A	5296	6644	7187	51
H68A	4453	6387	7922	55
H67A	4531	5557	8175	62
H66A	5452	4982	7693	64
H65A	6295	5239	6958	57
H70	1637	5950	3571	52
H71A	5681	9786	4006	133
H71B	6830	9967	4342	133
H71C	6395	9414	4405	133
H72A	6663	9261	3514	76
H72B	7169	9804	3481	76
H73A	8189	9060	4162	61
H73B	8706	9596	4093	61
H74A	8437	8880	3270	62
H74B	9019	9407	3228	62
H75A	10382	8801	3362	75
H75B	9915	8636	3897	75

H75C	10496	9165	3860	75
H71D	6039	9771	4529	146
H71E	7316	9825	4802	146
H71F	6730	9293	4752	146
H72C	7184	9860	3856	75
H72D	6694	9310	3829	75
H73C	8931	9565	4300	66
H73D	8441	9015	4273	66
H74C	8340	9030	3334	68
H74D	8830	9580	3361	68
H75D	10277	9003	3244	89
H75E	10090	8741	3788	89
H75F	10579	9291	3791	89
H71G	9372	9594	3620	68
H76A	9190	4983	1748	119
H76B	8092	5085	2035	119
H76C	8934	5528	1943	119
H77A	7724	5015	1072	92
H77B	8380	5527	1042	92
H78A	6992	5927	1489	66
H78B	6338	5414	1519	66
H79A	6582	5919	551	91
H79B	5926	5407	582	91
H80A	4855	6228	518	112
H80B	5102	6193	1148	112
H80C	4393	5768	821	112
H77C	7120	5338	1405	85
H77D	3974	6000	696	87
H77E	6305	6259	1086	79

Table S74. Atomic Occupancy for Gellman157.

Atom	***Occupancy***	**Atom**	***Occupancy***	**Atom**	***Occupancy***
O9	0.893(10)	C63	0.893(10)	H63A	0.893(10)
H63B	0.893(10)	C64	0.893(10)	C65	0.893(10)
H65	0.893(10)	C66	0.893(10)	H66	0.893(10)
C67	0.893(10)	H67	0.893(10)	C68	0.893(10)
H68	0.893(10)	C69	0.893(10)	H69	0.893(10)
O9A	0.107(10)	C63A	0.107(10)	H63C	0.107(10)
H63D	0.107(10)	C64A	0.107(10)	C69A	0.107(10)
H69A	0.107(10)	C68A	0.107(10)	H68A	0.107(10)
C67A	0.107(10)	H67A	0.107(10)	C66A	0.107(10)
H66A	0.107(10)	C65A	0.107(10)	H65A	0.107(10)
C71	0.687(3)	H71A	0.687(3)	H71B	0.687(3)
H71C	0.687(3)	C72	0.687(3)	H72A	0.687(3)
H72B	0.687(3)	C73	0.687(3)	H73A	0.687(3)
H73B	0.687(3)	C74	0.687(3)	H74A	0.687(3)
H74B	0.687(3)	C75	0.687(3)	H75A	0.687(3)
H75B	0.687(3)	H75C	0.687(3)	C71A	0.155(3)

H71D	0.155(3)	H71E	0.155(3)	H71F	0.155(3)
C72A	0.155(3)	H72C	0.155(3)	H72D	0.155(3)
C73A	0.155(3)	H73C	0.155(3)	H73D	0.155(3)
C74A	0.155(3)	H74C	0.155(3)	H74D	0.155(3)
C75A	0.155(3)	H75D	0.155(3)	H75E	0.155(3)
H75F	0.155(3)	Cl4B	0.1582(19)	Cl6B	0.1582(19)
Cl5B	0.1582(19)	C71B	0.1582(19)	H71G	0.1582(19)
C76	0.559(2)	H76A	0.559(2)	H76B	0.559(2)
H76C	0.559(2)	C77	0.559(2)	H77A	0.559(2)
H77B	0.559(2)	C78	0.559(2)	H78A	0.559(2)
H78B	0.559(2)	C79	0.559(2)	H79A	0.559(2)
H79B	0.559(2)	C80	0.559(2)	H80A	0.559(2)
H80B	0.559(2)	H80C	0.559(2)	Cl7A	0.2213(17)
Cl8A	0.2213(17)	Cl9A	0.2213(17)	C77A	0.2213(17)
H77C	0.2213(17)	Cl7B	0.1463(17)	Cl8B	0.1463(17)
Cl9B	0.1463(17)	C77B	0.1463(17)	H77D	0.1463(17)
Cl9C	0.0738(17)	Cl7C	0.0738(17)	Cl8C	0.0738(17)
C77C	0.0738(17)	H77E	0.0738(17)		