Electronic Supplementary Material (ESI) for Chemical Science.

Supplementary Information for
A Crystal-Structural Study of Pauling-Corey Rippled Sheets

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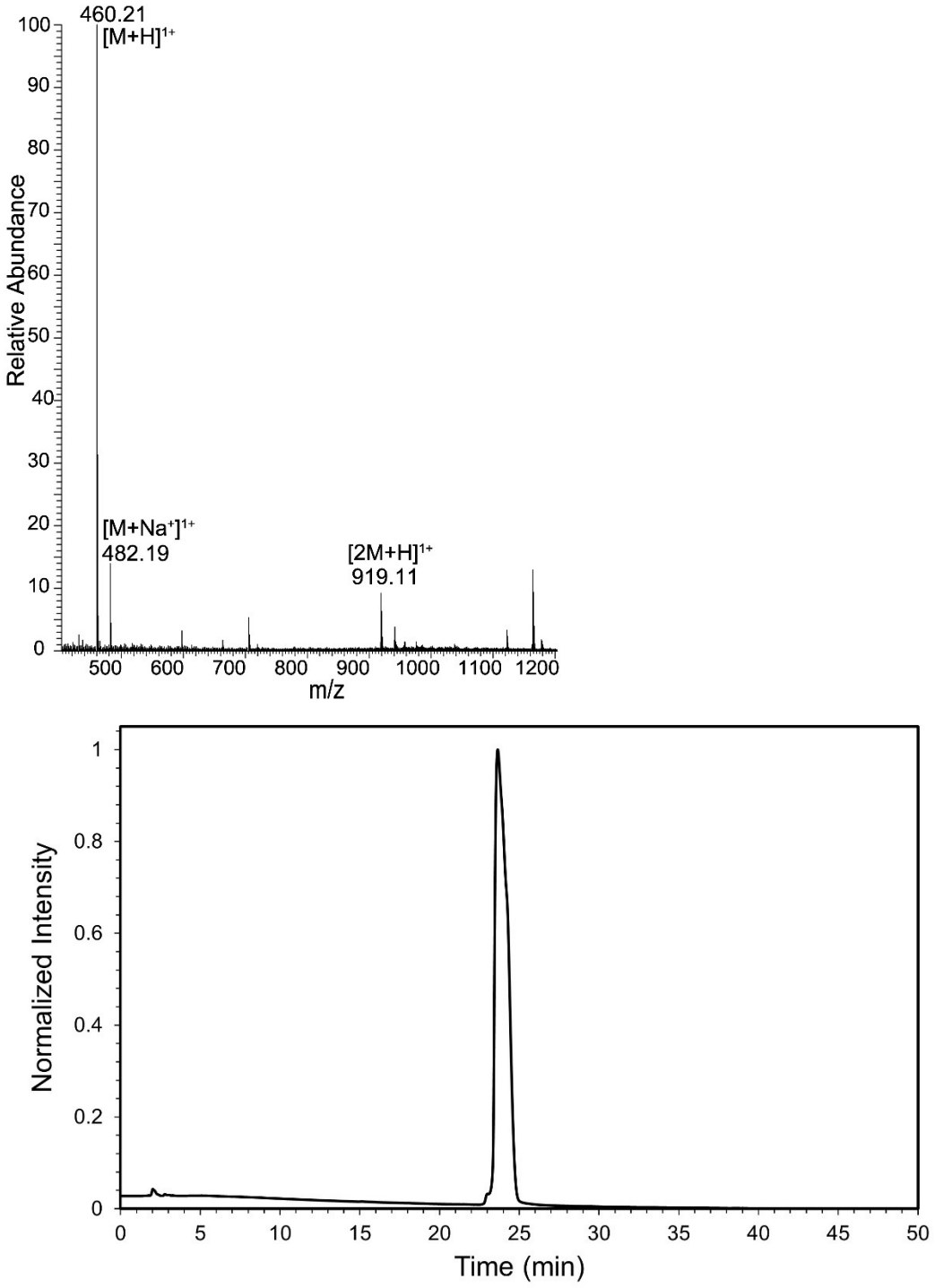

Fig. S1. Mass Spectrometry and HPLC characterization for NH_2-L-FFF-COOH.

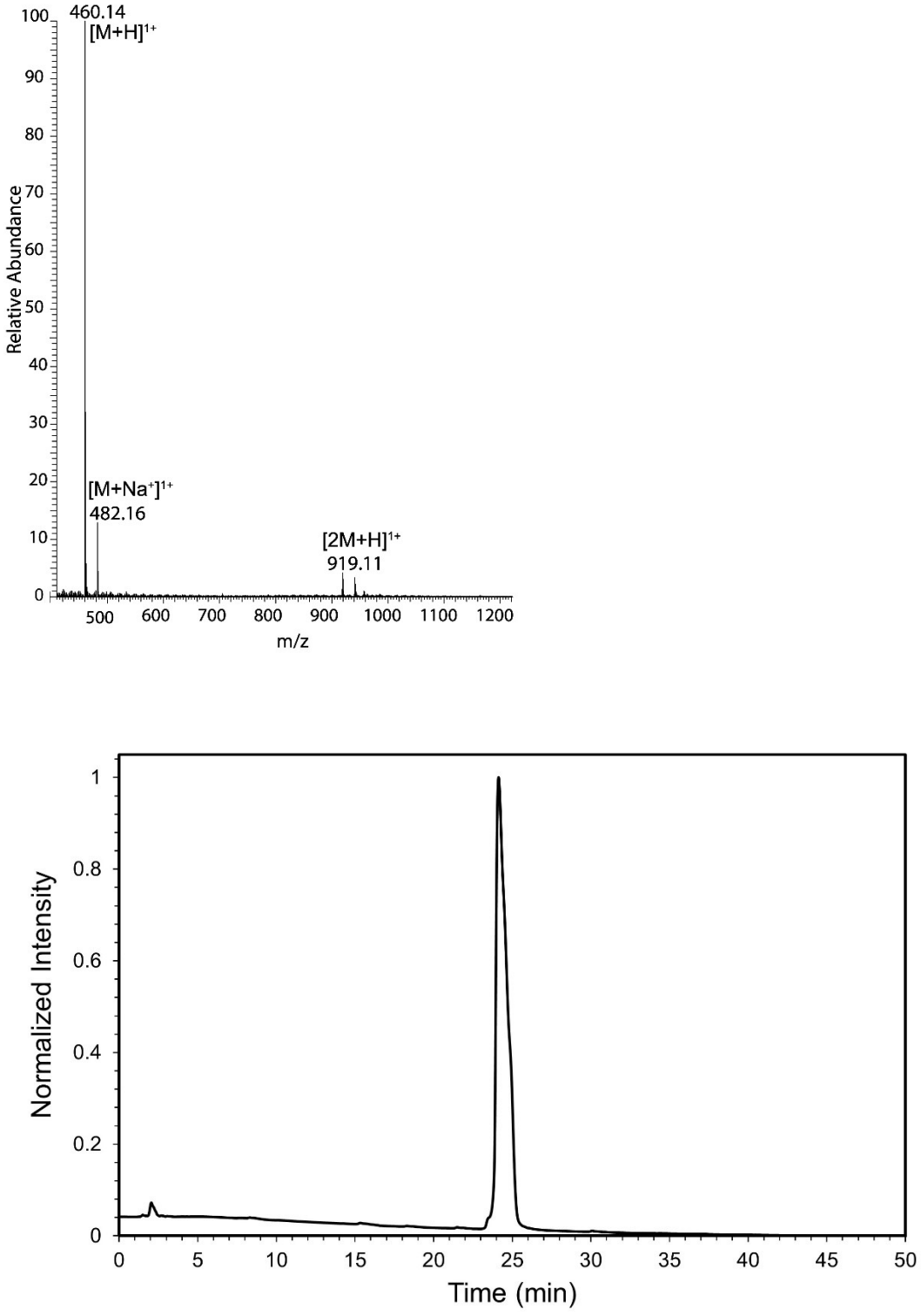

Fig. S2. Mass Spectrometry and HPLC characterization for NH_2-D-FFF-COOH.

Table S1. Crystallographic parameters

Compound	**Racemic triphenylalanine**
Formula	$C_{27}H_{29}N_3O_4$
FW	459.53
T (K)	100(2)
λ (Å)	1.54184
Crystal System	Monoclinic
Space group	$P2_1/c$
a (Å)	11.3563(5)
b (Å)	16.7472(6)
c (Å)	12.6545(6)
β (°)	101.483(4)
Volume (Å^3)	2358.54(18)
Z	4
ρ_{calc} (Mg/m^3)	1.294
Size (mm^3)	0.10 × 0.09 × 0.03
θ range (°)	4.44 – 67.06
Total data	23314
Unique data	4187
Parameters	323
Completeness	99.7%
R_{int}	5.00%
R_1 (I > 2σ)	4.43%
R_1 (all data)	5.54%
wR_2 (I > 2σ)	11.22%
wR_2 (all data)	11.81%
S	1.036
Min, max (e Å^{-3})	–0.279, 0.295

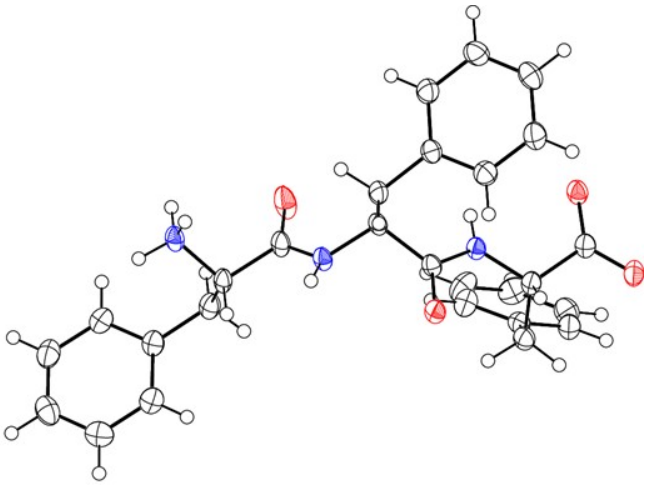

Fig. S3. Molecular graph of FFF with 50% thermal ellipsoids and H atoms shown as spheres of arbitrary radius. Color code: O red, N blue, C grey, H white.

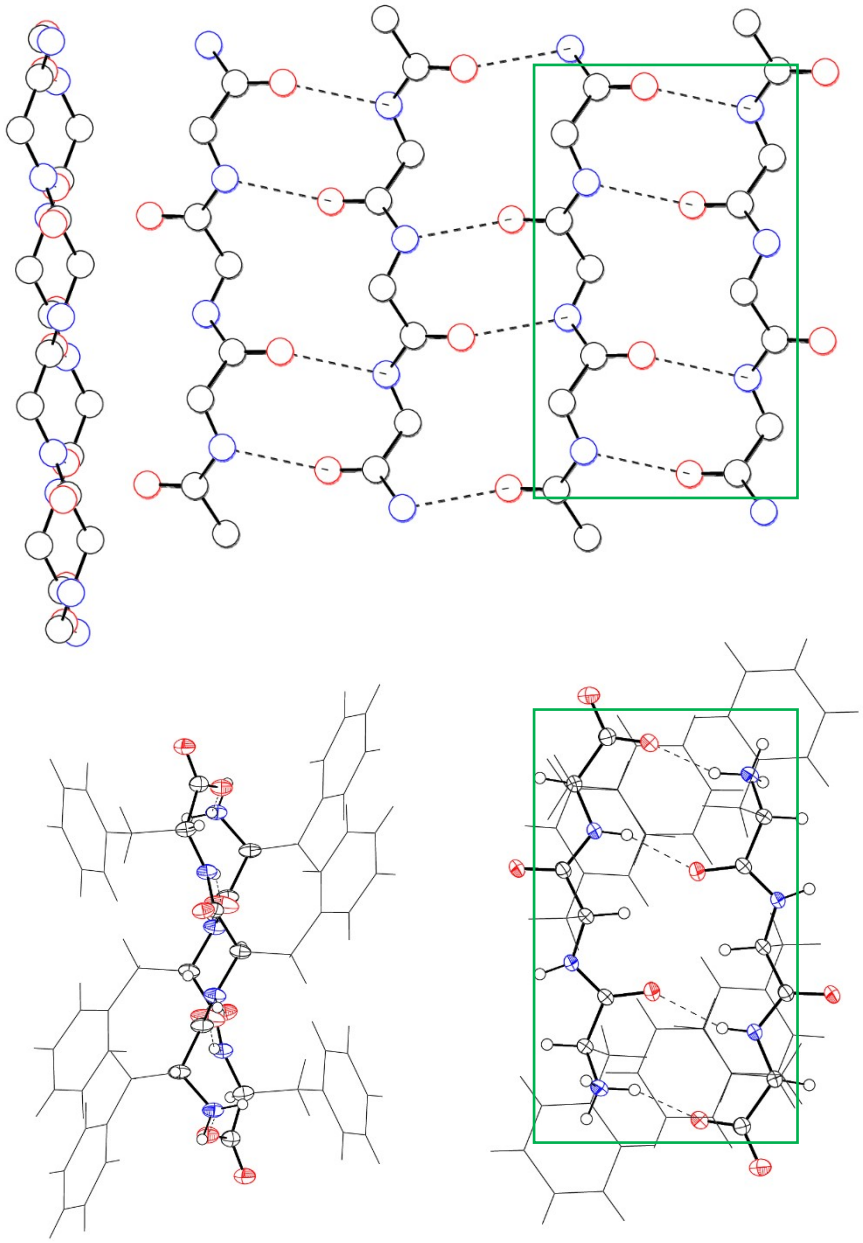

Fig. S4. *Top:* Side and top views of the rippled antiparallel sheet structure predicted by Pauling and Corey [cf. Figure 1 from *PNAS* **1953**, *39*, 253]. *Bottom:* Side and top views of the rippled sheet structure formed by FFF:fff. Atoms of the polypeptide backbone are shown as ellipsoids (non-H) or spheres of arbitrary radius (H). Side-chain atoms are shown as sticks. Color code: O red, N blue, C grey, H white. Hydrogen bonds are shown as dashed lines. Boxed in green is the similar motif from the predicted (top) and experimentally observed (bottom) structure.

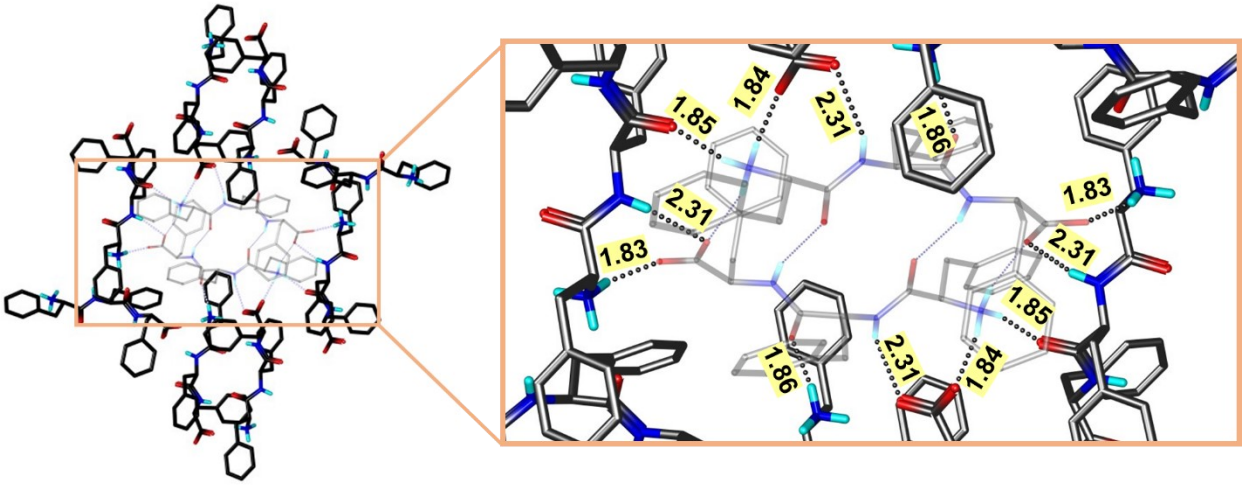

Fig. S5. The rippled antiparallel FFF:fff cross-β dimer shown in contact with its direct neighbors.

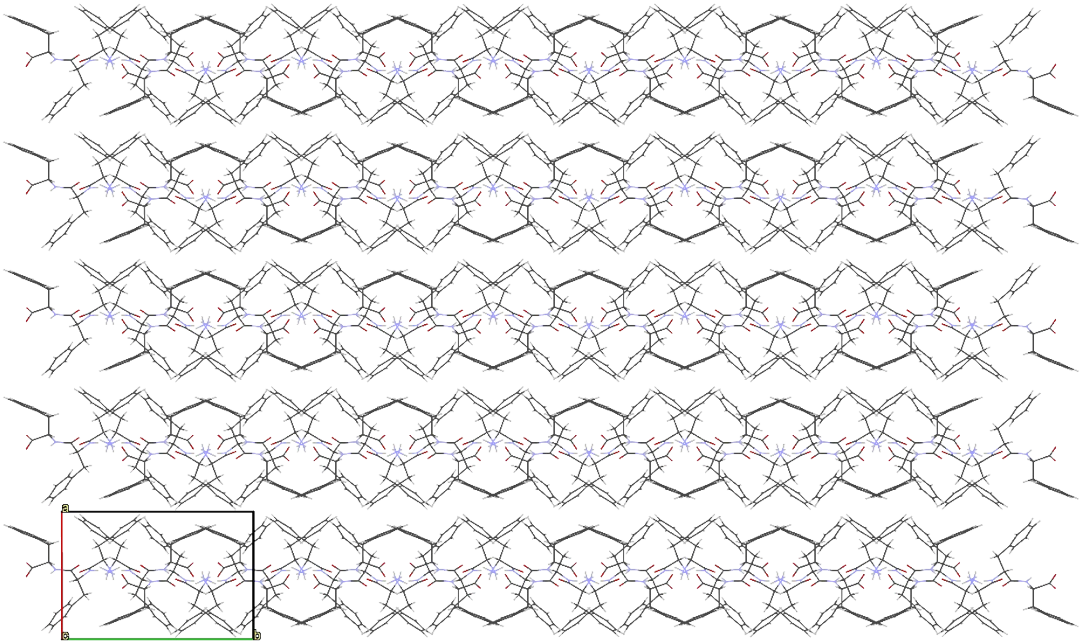

Fig. S6. Packing diagram of FFF:fff highlighting that the layer-to-layer distance is the crystallographic lattice parameter *a* = 11.3563(5) Å.

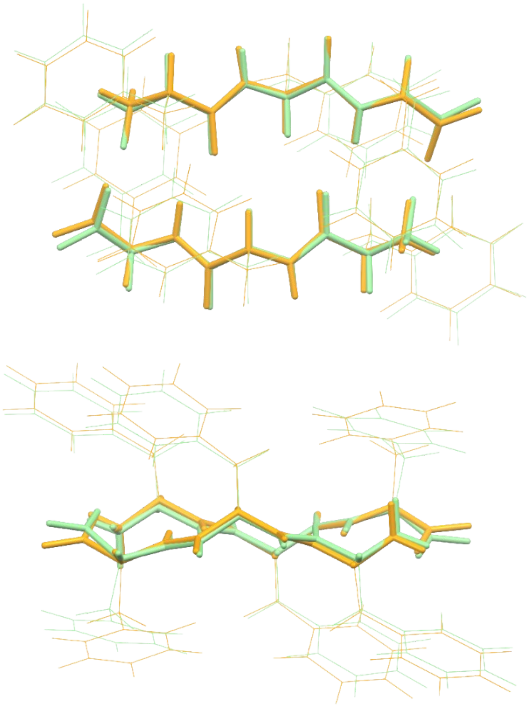

Fig. S7. Overlay of the crystallographic (orange) and the optimized (green; BP86-D3BJ/def2-SVP-CPCM$_{water}$) structure of the FFF:fff dimer. The polypeptide backbone is shown as thick sticks and the sidechains as thin sticks.

Table S2. Cartesian coordinates (Å) of the optimized (BP86-D3BJ/def2-SVP-CPCM$_{water}$) structure of FFF:fff.

N	4.09370415118074	12.55158927464286	8.01951364412157
H	3.96372050731073	11.87945342785070	8.89432214522468
H	3.67631800795364	13.45649622333425	8.29311390077636
H	5.10684826815001	12.72465726857804	7.86526886151337
O	4.68579699986856	10.00514061707158	7.08428367684645
C	4.21448263423020	10.83662052651225	6.28704928091444
C	3.40980475436177	12.02854135266891	6.80005942484666
H	3.38849381951550	12.83743527898451	6.04322345172614
C	1.97814220013488	11.57525758215830	7.15655122642748
H	1.54884004221062	11.10109355234939	6.25346523955252
H	2.05396185690734	10.79648718941633	7.94155838244508
C	1.09701622979355	12.71237139894564	7.62055690280438
C	1.09001748865928	13.10851995683912	8.97692250143198
H	1.71128992818933	12.55279025588311	9.70001424570751
C	0.29689600869346	14.19002383941204	9.39669431116368
H	0.30059192960416	14.48984062825498	10.45650801618807
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H	-1.12640255974232	15.72951255555404	8.80337394705965
C	-0.50629168506993	14.49105974185670	7.12126056936911
H	-1.13393642246107	15.02638237004613	6.39177196518855
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N	4.34294104144003	10.71642127776430	4.95139847892585
H	4.02617325620660	11.45980460038944	4.32051429068197
O	4.39183936657579	10.13695537762992	2.13900493459608
C	4.53310543154521	9.25649096794045	3.00946511222600
C	5.11461928813669	9.61794373703439	4.38145018136459
H	5.03142864014147	8.76479723071096	5.07907335395468
C	6.60638929528331	10.03204527673807	4.24216281816270
H	6.66557687589099	10.81001469482846	3.45165198777844
H	6.89946224238773	10.50545984207140	5.19887788497200
C	7.54254308228714	8.88459583473587	3.94395032674684
C	7.58844472576498	8.28365432032168	2.66674556186903
H	6.94541458912158	8.66743223090499	1.86030598840700
C	8.45406241058803	7.20893971981611	2.40909750515203
H	8.45866869918136	6.74144750394299	1.41303033853607
C	9.30756466458947	6.72918695577871	3.41817773094835
H	9.98108783486585	5.88275984854788	3.21748271978528
C	9.27910482335856	7.32382914438217	4.69135159978216
H	9.93367021228568	6.94739162654751	5.49237800535179
C	8.39410679692009	8.38361619037178	4.95215142881515
H	8.36419766813895	8.83646995965917	5.95638546896319
N	4.27663118712145	7.94432643547798	2.83632745682123
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O	5.09304882803972	5.85257505134016	0.05486234185960
C	4.90057255794537	6.14531097208455	1.25187163274656
C	3.97434193707562	7.35780504501355	1.54693871462719
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H	4.86982114067797	4.86859999022297	3.51236735404479
H	5.16572951866549	3.29077403479728	4.10731414188434
H	3.73588967731612	4.01485485660771	4.54727049323470
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C	4.63668287937818	5.91001586461808	6.11657509188658
C	5.43805826522110	4.71658749571353	5.60216894786604
H	5.46305755532070	3.90877021067308	6.35997761795219
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C	7.74531512969249	4.02995894798071	4.76861216913461
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C	4.30897502190871	7.49022987865021	9.39504697506979
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H	2.17747246094972	5.93349904559341	8.93207273703793
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C	0.37671889880669	9.52946860213518	9.97359028393214
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C	-0.47041059278953	10.01046241701172	8.95970510299747
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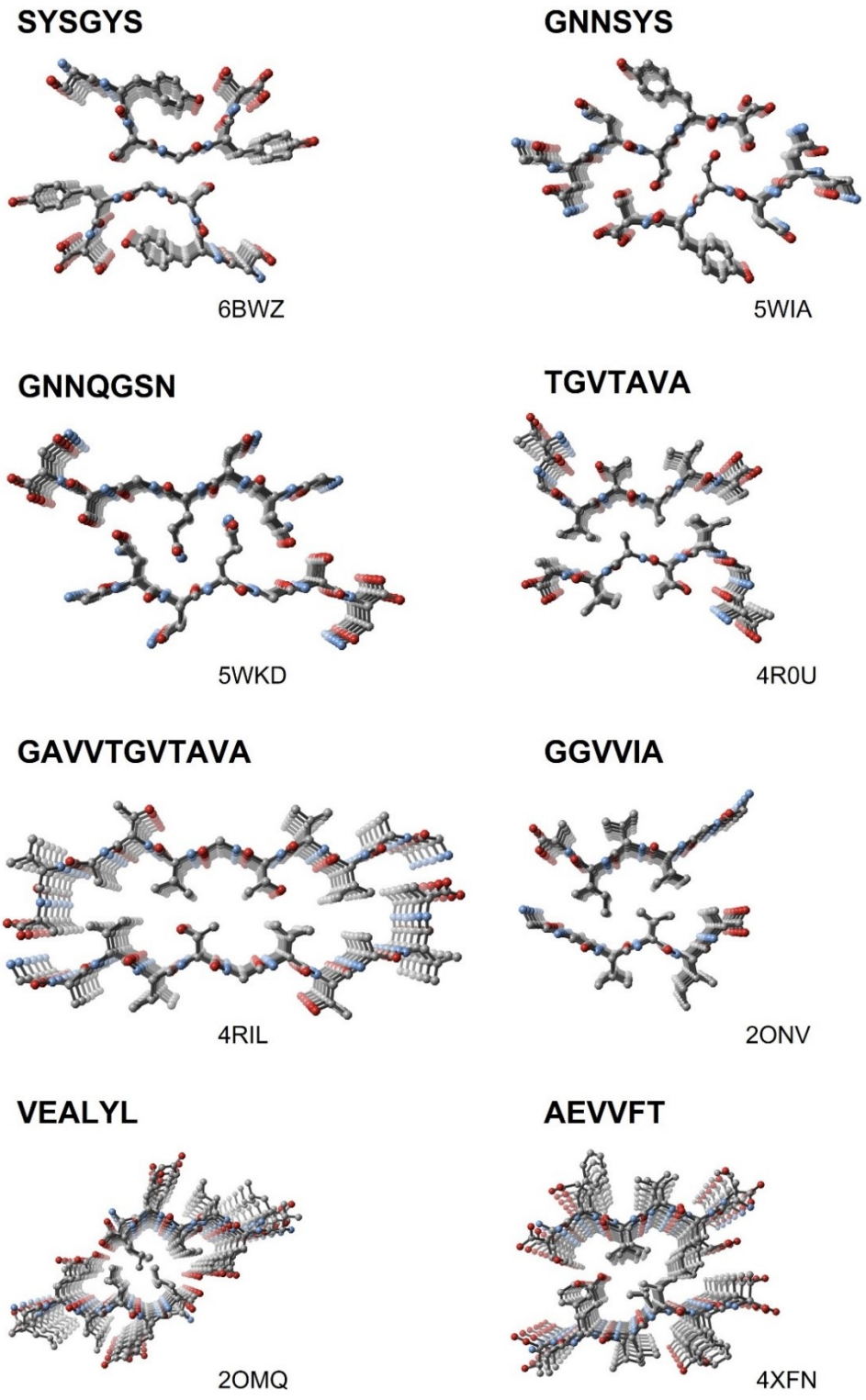

Fig. S8. Selected examples from Eisenberg and coworkers,(1–6) of fibrils formed by aggregating enantiopure peptides in which pleated β-sheets display one-dimensional, long-range order. Color code: C, gray; O, red; N, blue. Water molecules and hydrogens are omitted for clarity. PDB codes are displayed on the bottom right of each structure.

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