Supplementary material

The comparison of the commonly used B3LYP DFT functional and the new M05-2X DFT functional shows, that there are no significant changes in the geometries obtained. The only notable difference is the B3LYP optimized structure of the (For-Ala-NH_2)$_2$ dimer with 10-membered H-bonded ring in which the two strands adopt γ-turn conformation because of the weaker interaction energy predicted by the B3LYP compared to the M05-2X method. However, as the M05-2X functional accounts for dispersion forces, the predicted stabilities are greater compared to those of predicted by the B3LYP functional. As the counterpoise corrected values show, the BSSE error is around 3-4 kcal.mol^{-1} / residue thus it was necessary to apply this method for the analysis of sheets of different length.

STable 1. Backbone torsional angle values of antiparallel β-pleated sheet arrangements of For-(Ala)$_n$-NH_2 (n=1-6) as obtained at the M05-2X/6-31G(d) level of theory.

Type[a]	H-bonds	strand	φ1	ψ1	φ2	ψ2	φ3	ψ3	φ4	ψ4	φ5	ψ5	φ6	ψ6
S14	2	A	-158.1	162.2	-	-	-	-	-	-	-	-	-	-
		B	-158.2	162.4	-	-	-	-	-	-	-	-	-	-
S10	2	A	-160.4	162.9	-	-	-	-	-	-	-	-	-	-
		B	-160.4	162.9	-	-	-	-	-	-	-	-	-	-
S10/S14	3	A	-159.7	163	-160.9	157.3	-	-	-	-	-	-	-	-
		B	-163.1	167.9	-153.3	160.9	-	-	-	-	-	-	-	-
S14	4	A	-158.6	161.9	-161.4	163.5	-154.9	161.9	-	-	-	-	-	-
		B	-159.2	162.7	-161.7	164.2	-155.7	162.9	-	-	-	-	-	-
S10	4	A	-163.2	168.8	-155.1	161.5	-160.7	157.7	-	-	-	-	-	-
		B	-163.2	168.8	-155.1	161.5	-160.7	157.7	-	-	-	-	-	-
S10/S14	5	A	-163.5	168.3	-155.9	161.7	-162.2	163.1	174	-153.9	-	-	-	-
		B	-159.3	162.8	-162	163.9	-156.5	162.4	-160.6	157	-	-	-	-
S14	6	A	-159.4	162.8	-162	163.6	-156.7	162.6	-161.9	163.3	-154.4	161.6	-	-
		B	-159.4	162.8	-162	163.6	-156.7	162.6	-161.9	163.3	-154.4	161.6	-	-
S10	6	A	-163.4	168.1	-155.6	162.2	-162.2	163.5	-155.5	162	-159.9	156.8	-	-
		B	-163.4	168.1	-155.6	162.2	-162.2	163.5	-155.5	161.9	-159.9	156.8	-	-
S10/S14	7	A	-163.5	167.8	-155.7	162.3	-162.3	163.1	-156.5	162.2	-162.1	163	-154.1	161.5
		B	-159	163	-162.1	163.4	-156.3	162.6	-161.9	163.4	-155.5	162.4	-160	156.6

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 2. Backbone torsional angle values of antiparallel β-pleated sheet arrangements of For-$(Ala)_n$-NH_2 (n=1-6) as obtained at the B3LYP/6-31G(d) level of theory.

Type[a]	H-bonds	strand	$\varphi 1$	$\psi 1$	$\varphi 2$	$\psi 2$	$\varphi 3$	$\psi 3$	$\varphi 4$	$\psi 4$	$\varphi 5$	$\psi 5$	$\varphi 6$	$\psi 6$
S14	2	A	-151.6	153.7	-	-	-	-	-	-	-	-	-	-
		B	-151.6	153.7	-	-	-	-	-	-	-	-	-	-
S10	2	A	-80.4	80.5	-	-	-	-	-	-	-	-	-	-
		B	-80.4	80.6	-	-	-	-	-	-	-	-	-	-
S10/S14	3	A	-151.3	154.5	-152.4	144.6	-	-	-	-	-	-	-	-
		B	-157	160.2	-145.8	155.6	-	-	-	-	-	-	-	-
S14	4	A	-152.1	155.1	-157.6	156.4	-147	155.8	-	-	-	-	-	-
		B	-152.1	155.1	-157.6	156.4	-147	155.9	-	-	-	-	-	-
S10	4	A	-156.2	159.3	-145.3	154.1	-151.2	145.1	-	-	-	-	-	-
		B	-156.2	159.3	-145.4	154.1	-151.2	145	-	-	-	-	-	-
S10/S14	5	A	-156.8	158.7	-147.1	155.3	-157.7	156.4	-147.4	156.3	-	-	-	-
		B	-152	155.3	-157.2	156.4	-148.4	154.3	-150.9	143.9	-	-	-	-
S14	6	A	-151.9	155.2	-157.4	156.2	-148.8	156.2	-157.2	155.8	-147.5	155.9	-	-
		B	-151.9	155.2	-157.4	156.2	-148.8	156.2	-157.2	155.8	-147.5	155.9	-	-
S10	6	A	-156.8	158.5	-147	154.7	-157.2	155.6	-147.4	153.9	-151	144.4	-	-
		B	-156.8	158.5	-147	154.8	-157.2	155.6	-147.4	153.9	-151	144.5	-	-
S10/S14	7	A	-156.8	158.3	-147.4	154.8	-157.5	155.7	-148.2	155.9	-157.3	156.5	-147.7	156.5
		B	-151.7	155.2	-157.4	156.1	-148.9	156	-156.8	155.5	-148	153.9	-151.3	144.5

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 3. Thermodynamic functions of antiparallel β-pleated sheet arrangements of For-$(Ala)_n$-NH_2 (n=1-6) as obtained at various levels of theory

Type[a]	H-bonds	B3LYP/6-31G(d)			M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
S14	2	-834.188	-834.258	147.25	-834.064	-834.131	140.559	-834.337	-834.404	147.25
S10	2	-834.173	-834.249	160.153	-834.047	-834.118	151.186	-834.321	-834.393	160.153
S10/S14	3	-1328.67	-1328.77	208.98	-1328.48	-1328.58	199.07	-1328.91	-1328.99	208.98
S14	4	-1823.16	-1823.28	257.11	-1822.92	-1823.04	244.88	-1823.49	-1823.61	257.11
S10	4	-1823.14	-1823.27	267.11	-1822.90	-1823.03	256.81	-1823.47	-1823.59	267.11
S10/S14	5	-2317.64	-2317.79	318.23	-2317.34	-2317.49	301.58	-2318.06	-2318.2	318.23
S14	6	-2812.15	-2812.33	373.71	-2811.78	-2811.95	348.64	-2812.64	-2812.81	373.71
S10	6	-2812.13	-2812.32	386.25	-2811.76	-2811.93	361.78	-2812.63	-2812.8	386.25
S10/S14	7	-3306.61	-3306.82	429.97	-3306.2	-3306.39	408.23	-3307.21	-3307.41	429.97

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 4. Thermodynamic functions of antiparallel β-pleated sheet arrangements of For-$(Ala)_n$-NH_2 (n=1-6) in water as obtained at the M05-2X/6-311++G(d,p)//M05-2X/6-31G(d) level of theory

Type[a]	H-bonds	M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
S14	2	-834.355	-834.422	147.25
S10	2	-834.35	-834.422	160.153
S10/S14	3	-1328.93	-1329.03	208.98
S14	4	-1823.52	-1823.64	257.11
S10	4	-1823.51	-1823.64	267.11
S10/S14	5	-2318.1	-2318.24	318.23
S14	6	-2812.68	-2812.85	373.71
S10	6	-2812.68	-2812.85	386.25
S10/S14	7	-3307.26	-3307.46	429.97

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 5. The change in thermodynamic functions during the dimerization of antiparallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) as obtained at various levels of theory

Type[a]	H-bonds	B3LYP/6-31G(d)			M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}	ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}	ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}
S14	2	-17.28	-4.89	-12.31	-20.93	-7.97	-12.96	-18.98	-6.02	-12.96
S10	2	-7.87	0.65	-8.47	-9.89	-0.10	-9.79	-8.66	1.14	-9.79
S10/S14	3	-17.54	-4.88	-12.58	-22.62	-8.88	-13.74	-20.46	-6.72	-13.74
S14	4	-28.29	-12.18	-16.11	-36.54	-18.98	-17.57	-33.13	-15.56	-17.57
S10	4	-17.43	-4.30	-13.13	-24.33	-10.32	-14.01	-21.88	-7.87	-14.01
S10/S14	5	-28.16	-11.60	-16.56	-37.91	-18.74	-19.17	-34.30	-15.13	-19.17
S14	6	-38.70	-18.95	-19.63	-51.64	-28.71	-22.93	-46.82	-23.89	-22.93
S10	6	-28.13	-12.14	-15.89	-39.46	-20.44	-19.01	-35.59	-16.58	-19.01
S10/S14	7	-38.45	-18.06	-20.39	-53.03	-28.67	-24.36	-47.99	-23.62	-24.36

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 6. The change in thermodynamic functions during the dimerization of antiparallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) in water as obtained at the M05-2X/6-311++G(d,p)//M05-2X/6-31G(d) level of theory

Type[a]	H-bonds	M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}
S14	2	-8.75	4.21	-12.96
S10	2	-5.30	4.49	-9.79
S10/S14	3	-11.72	2.02	-13.74
S14	4	-18.24	-0.68	-17.57
S10	4	-14.77	-0.76	-14.01
S10/S14	5	-21.28	-2.11	-19.17
S14	6	-27.80	-4.87	-22.93
S10	6	-24.46	-5.44	-19.01
S10/S14	7	-30.89	-6.53	-24.36

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even H-bond number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tripeptide, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd H-bond number there is only one possible structure indicated as S10/S14.

STable 7. Backbone torsional angle values of parallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) as obtained at the M05-2X/6-31G(d) level of theory.

Type[a]	H-bonds[b]	strand	φ1	ψ1	φ2	ψ2	φ3	ψ3	φ4	ψ4	φ5	ψ5	φ6	ψ6
S12	2	A	-160.9	165.1	-	-	-	-	-	-	-	-	-	-
		B	-82.6	73.6	-	-	-	-	-	-	-	-	-	-
S12	3	A	-155.8	-150.5	-161.4	163.9	-	-	-	-	-	-	-	-
		B	-158.2	147.1	-82.2	76.3	-	-	-	-	-	-	-	-
S12	4	A	-153.2	160.5	-162.6	-164.4	-159.5	164.3	-	-	-	-	-	-
		B	-159	159.7	-148.9	141.6	-83.1	77.1	-	-	-	-	-	-
S12	5	A	-162.5	167.9	-150.7	160.4	-160.2	-165.7	-159.2	164	-	-	-	-
		B	-157.5	160.9	-159.6	162.5	-151.7	144.5	-83.4	76.7	-	-	-	-
S12	6*	A	-159.8	155.2	-156	160.6	-159.5	166.2	-153	145.2	-83.1	77.4	-	-
		B	-155.9	162.9	-161.6	164.1	-153.1	162.8	-158.6	-162.8	-159.7	164.4	-	-
S12	7*	A	-162.2	166.1	-154	162.7	177.8	-160.4	-152.4	162.5	-158.6	-162.9	-159.8	164.1
		B	-156.3	163.4	-157.5	153	-157.1	162.3	-158.7	166	-153.6	145.6	-83	77.5

[a] S12: parallel sheets containing 12-membered H-bonded rings.

[b] *: structures with 1 imaginary frequency. This partial optimization was found to be sufficient as the error in entropy values obtained is smaller than 0.5 kcal.mol^{-1}.

STable 8. Thermodynamic functions of parallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) as obtained at various levels of theory

Type[a]	H-bonds[b]	M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
S12	2	-834.064	-834.13	138.474	-834.335	-834.401	138.474
S12	3	-1328.48	-1328.58	191.39	-1328.9	-1328.99	191.39
S12	4	-1822.91	-1823.03	248.15	-1823.48	-1823.6	248.15
S12.	5	-2317.34	-2317.48	302.289	-2318.05	-2318.2	302.289
S12	6*	-2811.77	-2811.93	348.03	-2812.63	-2812.79	348.03
S12	7*	-3306.19	-3306.38	404.26	-3307.2	-3307.4	404.26

[a] S12: parallel sheets containing 12-membered H-bonded rings.

[b] *: structures with 1 imaginary frequency. This partial optimization was found to be sufficient as the error in entropy values obtained is smaller than 0.5 kcal.mol^{-1}.

STable 9. Thermodynamic functions of parallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) in water as obtained at the M05-2X/6-311++G(d,p)//M05-2X/6-31G(d) level of theory

Type[a]	H-bonds	M05-2X/6-311++G(d,p)//M05-2X/6-31G(d)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
S12	2	-834.354	-834.42	138.474
S12	3	-1328.93	-1329.02	191.39
S12	4	-1823.51	-1823.63	248.15
S12.	5	-2318.09	-2318.24	302.289
S12	6*	-2812.67	-2812.84	348.03

[a] S12: parallel sheets containing 12-membered H-bonded rings.

[b] *: structures with 1 imaginary frequency. This partial optimization was found to be sufficient as the error in entropy values obtained is smaller than 0.5 kcal.mol^{-1}

STable 10. The averaged values of antiparallel β-pleated sheet structures of For-(Ala)$_n$-NH$_2$ (n=1-6) and the change in thermodynamic functions during the dimerization of parallel β-pleated sheet arrangements of For-(Ala)$_n$-NH$_2$ (n=1-6) as obtained at various levels of theory.

Type[a]	H-bonds[b]	M05-2X/6-311++G(d,p)//M05-2X/6-31G(d) vacuum			M05-2X/6-311++G(d,p)//M05-2X/6-31G(d) water		
		Δ*H*/ kcal.mol^{-1}	Δ*G*/ kcal.mol^{-1}	*T*Δ*S*/ kcal.mol^{-1}	Δ*H*/ kcal.mol^{-1}	Δ*G*/ kcal.mol^{-1}	*T*Δ*S*/ kcal.mol^{-1}
S12	2	-17.67	-4.09	-13.58	-7.98	5.60	-13.58
S12	3	-19.80	-3.77	-16.03	-9.92	6.11	-16.03
S12	4	-24.92	-8.33	-16.59	-13.17	3.42	-16.59
S12.	5	-30.61	-11.66	-18.96	-17.60	1.36	-18.96
S12	6*	-36.34	-13.22	-23.11	-21.49	1.63	-23.11
S12	7*	-42.11	-16.57	-25.55	-	-	-
Avg.	2	-13.82	-2.44	-11.37	-7.03	4.35	-11.37
Avg.	3	-20.46	-6.72	-13.74	-11.72	2.02	-13.74
Avg.	4	-27.50	-11.71	-15.79	-16.51	-0.72	-15.79
Avg.	5	-34.30	-15.13	-19.17	-21.28	-2.11	-19.17
Avg.	6	-41.21	-20.23	-20.97	-26.13	-5.16	-20.97
Avg.	7	-47.99	-23.62	-24.36	-30.89	-6.53	-24.36

[a] S12: parallel sheets containing 12-membered H-bonded rings. Avg.: Average of the values obtained for S10 and S14 (starting with 10- or 14-membered H-bonded system) antiparallel sheets with the same residue number.

[b] *: structures with 1 imaginary frequency. This partial optimization was found to be sufficient as the error in entropy values obtained is smaller than 0.5 kcal.mol^{-1}.

In the multiple stranded case, here is reported the comparison of the values obtained from the calculations at two levels of theory, M05-2X/6-31G(d) and M05-2X/6-311++G(d,p). The calculations carried out with the triple zeta basis set show lower stability for the sheets, in accord with the decreasing BSSE with the increasing size of the basis set.

STable 11. Thermodynamic functions of parallel β-pleated sheet arrangements of monoalanine strands as obtained at various levels of theory

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
-	1	-417.02	-417.06	92.01	-417.15	-417.20	92.01
S12	2	-834.06	-834.13	138.47	-834.34	-834.40	138.47
S12	3	-1251.10	-1251.20	192.58	-1251.51	-1251.60	192.58
S12.	4	-1668.14	-1668.26	244.75	-1668.69	-1668.80	244.75
S12	5	-2085.19	-2085.33	298.73	-2085.86	-2086.00	298.73
S12	6	-2502.23	-2502.39	349.75	-2503.04	-2503.20	349.75

[a] S12: parallel sheets containing 12-membered H-bonded rings

STable 12. Thermodynamic functions of antiparallel β-pleated sheet arrangements of monoalanine strands as obtained at various levels of theory

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311G++(d,p)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
S14	2	-834.05	-834.12	151.19	-834.32	-834.39	151.19
S10	2	-834.06	-834.13	140.56	-834.34	-834.40	140.56
S10/S14	3	-1251.10	-1251.19	199.78	-1251.51	-1251.60	199.78
S14	4	-1668.13	-1668.25	259.72	-1668.68	-1668.80	259.72
S10	4	-1668.15	-1668.27	245.97	-1668.69	-1668.81	245.97
S10/S14	5	-2085.18	-2085.33	305.92	-2085.86	-2086.01	305.92
S14	6	-2502.22	-2502.39	364.80	-2503.03	-2503.21	364.80
S10	6	-2502.24	-2502.40	351.68	-2503.05	-2503.22	351.68

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even strand number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tetramer, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd strand number there is only one possible structure indicated as S10/S14.

STable 13. The averaged values of antiparallel β-pleated sheet structures of monoalanine strands and the change in thermodynamic functions during the oligomerization of parallel β-pleated sheet arrangements of monoalanine strands as obtained at various levels of theory.

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)		
		ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}	ΔH/ kcal.mol^{-1}	ΔG/ kcal.mol^{-1}	$T\Delta S$/ kcal.mol^{-1}
Avg.	2	-7.71	-2.02	-5.69	-6.91	-1.22	-5.69
Avg.	3	-10.74	-3.16	-7.58	-9.67	-2.09	-7.58
Avg.	4	-12.36	-3.77	-8.59	-11.11	-2.52	-8.59
Avg.	5	-13.33	-4.14	-9.19	-12.01	-2.81	-9.19
Avg.	6	-13.97	-4.34	-9.63	-12.57	-2.94	-9.63
S12	2	-10.34	-3.55	-6.79	-8.83	-2.04	-6.79
S12	3	-12.06	-3.77	-8.29	-10.59	-2.29	-8.29
S12	4	-12.99	-3.80	-9.19	-11.37	-2.18	-9.19
S12	5	-13.55	-3.93	-9.62	-11.87	-2.25	-9.62
S12	6	-13.93	-3.87	-10.05	-12.21	-2.16	-10.05

[a] S12: parallel sheets containing 12-membered H-bonded rings. Avg.: Average of the values obtained for S10 and S14 (starting with 10- or 14-membered H-bonded system) antiparallel sheets with the same strand number.

STable 14. Thermodynamic functions of parallel β-pleated sheet arrangements of dialanine strands as obtained at various levels of theory

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)		
		H/ kcal.mol^{-1}	G/ kcal.mol^{-1}	S/ Cal.mol^{-1}K^{-1}	H/ kcal.mol^{-1}	G/ kcal.mol^{-1}	S/ Cal.mol^{-1}K^{-1}
-	1	-664.22	-664.28	122.58	-664.44	-664.44	122.58
S12	2	-1328.48	-1328.58	191.39	-1328.90	-1328.90	191.39
S12	3	-1992.74	-1992.87	275.43	-1993.36	-1993.36	275.43
S12.	4	-2657.00	-2657.17	350.75	-2657.84	-2657.84	350.75
S12	5	-3321.26	-3321.46	427.32	-3322.30	-3322.30	427.32
S12	6	-3985.51	-3985.76	514.55	-3986.77	-3986.77	514.55

[a] S12: parallel sheets containing 12-membered H-bonded rings

STable 15. Thermodynamic functions of antiparallel β-pleated sheet arrangements of dialanine strands as obtained at various levels of theory

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311G++(d,p)		
		H/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}	*H*/ kcal.mol^{-1}	*G*/ kcal.mol^{-1}	*S*/ Cal.mol^{-1}K^{-1}
-	1	-664.22	-664.28	122.58	-664.44	-664.49	122.58
S10/S14	2	-1328.48	-1328.58	199.07	-1328.90	-1329.00	199.07
S10/S14	3	-1992.75	-1992.88	275.64	-1993.38	-1993.51	275.64
S10/S14	4	-2657.01	-2657.18	354.48	-2657.85	-2658.02	354.48
S10/S14	5	-3321.27	-3321.47	427.79	-3322.32	-3322.52	427.79
S10/S14	6	-3985.53	-3985.78	507.67	-3986.79	-3987.03	507.67

[a] Antiparallel sheets contain alternating 10- and 14-membered H-bonded rings. In the case of even strand number, the sheet contains an uneven number of 10- and 14-membered, H-bonded systems resulting in 2 possible structures. S10 or S14 indicates the first H-bonded system in a given sheet (e.g. for a tetramer, S14 indicates the S14S10S14 system while S10 indicates the S10S14S10 system). For peptides with odd strand number there is only one possible structure indicated as S10/S14.

STable 16. The averaged values of antiparallel β-pleated sheet structures of the dialanine strands and the change in thermodynamic functions during the oligomerization of parallel β-pleated sheet arrangements of dialanine strands as obtained at various levels of theory.

Type[a]	Strands	M05-2X/6-31G(d)			M05-2X/6-311++G(d,p)		
		Δ*H*/ kcal.mol^{-1}	Δ*G*/ kcal.mol^{-1}	*T*Δ*S*/ kcal.mol^{-1}	Δ*H*/ kcal.mol^{-1}	Δ*G*/ kcal.mol^{-1}	*T*Δ*S*/ kcal.mol^{-1}
S10/S14	2	-11.31	-4.44	6.87	-10.23	-3.36	6.87
S10/S14	3	-15.48	-6.33	6.33	-13.99	-4.84	9.15
S10/S14	4	-17.77	-7.65	10.12	-16.06	-5.94	10.12
S10/S14	5	-19.12	-8.08	11.04	-17.28	-6.25	11.04
S10/S14	6	-20.02	-8.70	11.32	-18.05	-6.73	11.32
S12	2	-11.58	-3.57	8.01	-9.90	-1.89	8.01
S12	3	-13.54	-4.37	4.37	-11.78	-2.61	2.61
S12	4	-16.36	-5.96	10.40	-14.42	-4.02	10.40
S12	5	-17.42	-6.36	11.07	-15.26	-4.19	11.07
S12	6	-17.77	-6.79	10.98	-15.63	-4.65	10.98

[a] S12: parallel sheets containing 12-membered H-bonded rings. Avg.: Average of the values obtained for S10 and S14 (starting with 10- or 14-membered H-bonded system) antiparallel sheets with the same strand number.

STable 17. Correlation between the raw and normalized thermodynamic (Y) functions, vs. the number of inter–chain H–bonds (*n*). Fitting parameters obtained

(*Y*) of parallel	*Y* = m*n*+b		R^2	(*Y*/*n*) of parallel	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*TΔS*	2.56	7.27	0.946	-*TΔS*/*n*	2.28	8.59	0.893
ΔH	-5.60	-2.74	0.999	*ΔH*/*n*	-5.54	-3.04	0.956
ΔG	-3.04	4.53	0.975	*ΔG*/*n*	-3.26	5.54	0.847
(*Y*) of antiparallel	***Y* = m*n*+b**		R^2	**(*Y*/*n*) of antiparallel**	***Y*/*n* = M+B/*n***		R^2
	m	**b**			**M**	**B**	
-*TΔS*	2.57	5.99	0.993	-*TΔS*/*n*	2.50	6.30	0.990
ΔH	-6.85	-0.04	0.999	*ΔH*/*n*	-6.83	-0.09	0.187
ΔG	-4.28	5.95	0.997	*ΔG*/*n*	-4.34	6.21	0.989

STable 18. Correlation between the raw and normalized thermodynamic (Y) functions, vs. the number of inter–chain H–bonds *(n)*, in water. Fitting parameters obtained

(*Y*) of parallel	*Y* = m*n*+b		R^2	(*Y*/*n*) of parallel	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*TΔS*	2.56	6.56	0.896	-*TΔS*/*n*	2.28	7.87	0.876
ΔH	-3.91	2.06	0.997	*ΔH*/*n*	-3.84	1.75	0.744
ΔG	-1.55	10.1	0.838	*ΔG*/*n*	-1.80	11.2	0.951
(*Y*) of antiparallel	***Y* = m*n*+b**		R^2	**(*Y*/*n*) of antiparallel**	***Y*/*n* = M+B/*n***		R^2
	m	**b**			**M**	**B**	
-*TΔS*	2.57	5.99	0.987	-*TΔS*/*n*	2.50	6.30	0.990
ΔH	-4.78	2.59	0.999	*ΔH*/*n*	-4.77	2.53	0.999
ΔG	-2.21	8.58	0.991	*ΔG*/*n*	-2.56	8.83	0.996

STable 19. Correlation between the raw and normalized thermodynamic (Y) functions, vs. the number of peptide strands of monoalanine. Fitting parameters obtained

(*Y*) of parallel	*Y* = m*n*+b		R^2	(*Y*/*n*) of parallel	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*T*Δ*S*	11.67	-9.95	0.999	-*T*Δ*S*/*n*	11.60	-9.70	0.997
Δ*H*	-13.88	10.02	0.999	Δ*H*/*n*	-13.90	10.09	0.999
Δ*G*	-2.21	0.06	0.992	Δ*G*/*n*	-2.30	0.38	0.059
(*Y*) of antiparallel	*Y* = m*n*+b		R^2	(*Y*/*n*) of antiparallel	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*T*Δ*S*	11.61	-11.98	0.999	-*T*Δ*S*/*n*	11.55	-11.79	0.999
Δ*H*	-15.42	17.14	0.999	Δ*H*/*n*	-15.38	17.00	0.999
Δ*G*	-3.82	5.16	0.999	Δ*G*/*n*	-3.83	5.21	0.999

STable 20. Correlation between the raw and normalized thermodynamic (Y) functions, vs. the number of peptide strands of dialanine. Fitting parameters obtained

(*Y*) of monoalanine	*Y* = m*n*+b		R^2	(*Y*/*n*) of monoalanine	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*T*Δ*S*	11.61	-11.98	0.999	-*T*Δ*S*/*n*	11.55	-11.79	0.999
Δ*H*	-15.42	17.14	0.999	Δ*H*/*n*	-15.38	17.00	0.999
Δ*G*	-3.82	5.16	0.999	Δ*G*/*n*	-3.83	5.21	0.999
(*Y*) of dialanine	*Y* = m*n*+b		R^2	(*Y*/*n*) of dialanine	*Y*/*n* = M+B/*n*		R^2
	m	b			M	B	
-*T*Δ*S*	13.61	-13.47	0.999	-*T*Δ*S*/*n*	13.61	-13.47	0.997
Δ*H*	-22.01	23.76	0.999	Δ*H*/*n*	-21.95	23.54	0.999
Δ*G*	-8.40	10.29	0.999	Δ*G*/*n*	-8.34	10.07	0.991