Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

SUPPLEMENTARY INFORMATION

Interfacial Water at the Trialanine Hydrophilic Surface. A DFT Electronic Structure and Bottom-Up Investigation

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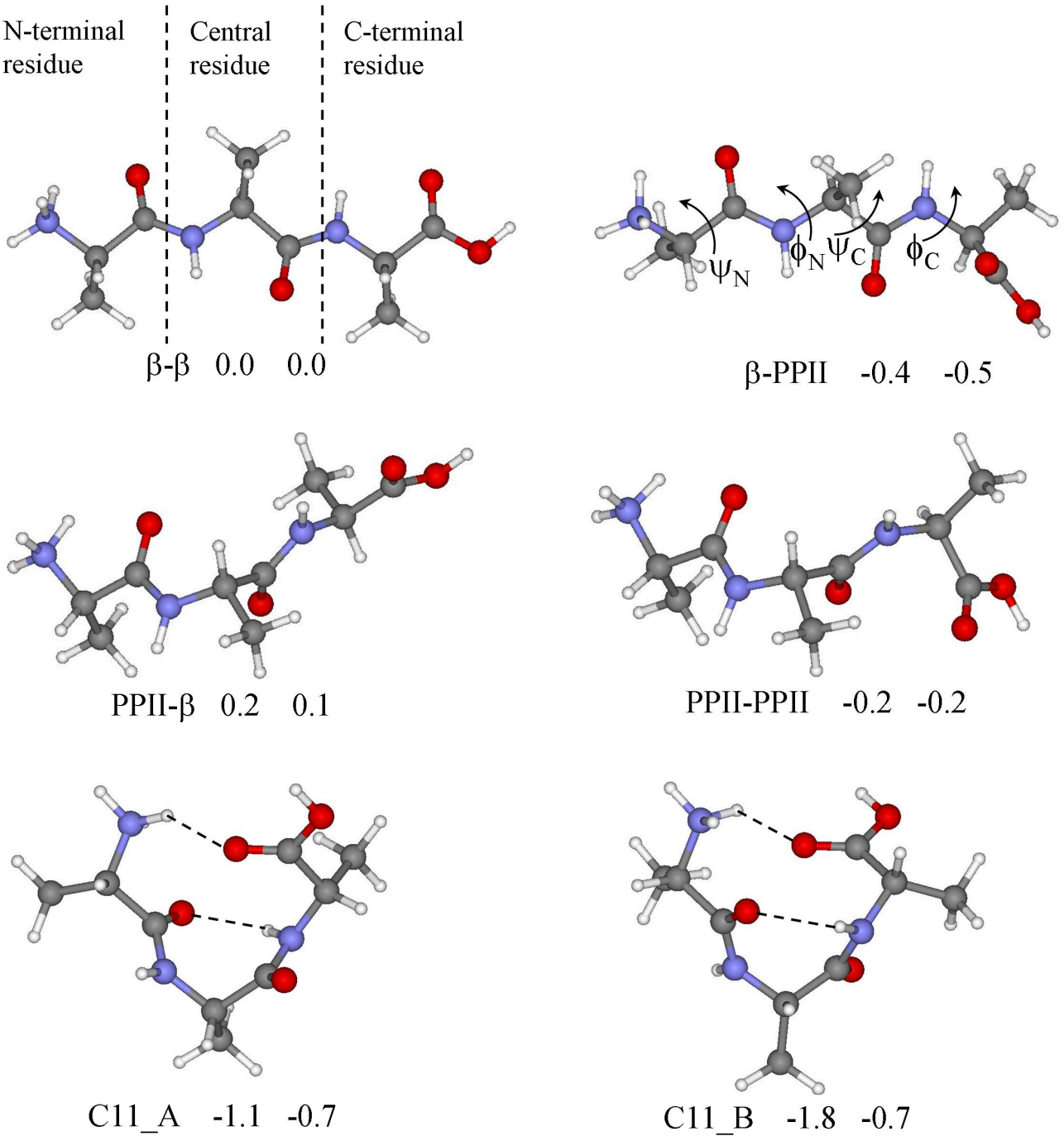

Fig. S1. Molecular structures and relative energy (kcal mol^{-1}) at the 6-31+G* (value on the left) and aug-cc-pVTZ (value on the right) of the "bare" $AlaH_3^+$ peptide in various conformations.

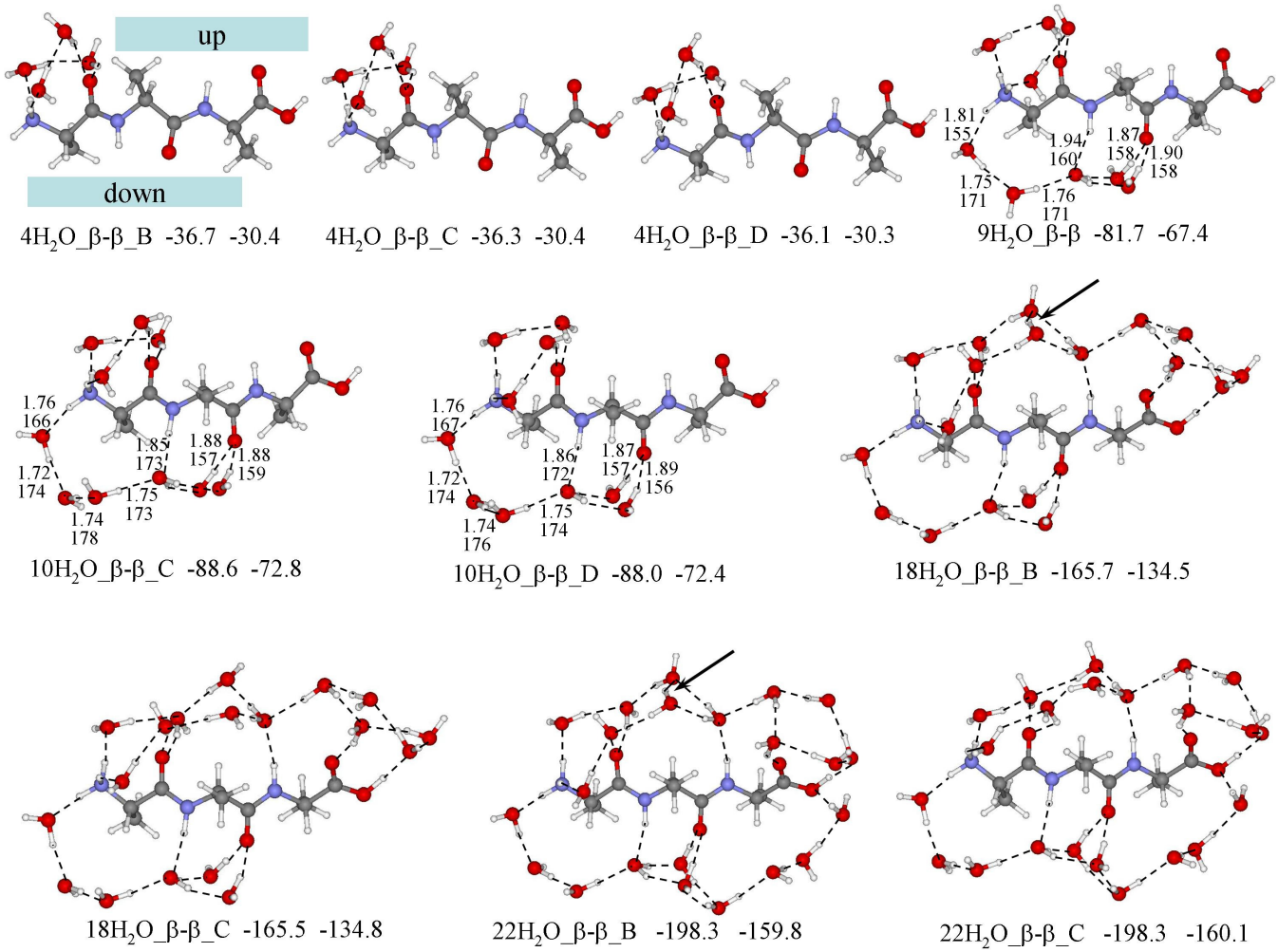

Fig. S2. Optimized molecular structures of $Ala_3H^+ \cdot nH_2O$ complexes with peptidic chain in the β-β conformation. Formation energies (kcal mol^{-1}) at the 6-31+G* (left value) and aug-cc-pVTZ (right value) have been computed relative to the isolated Ala_3H^+ (in the β-β conformation) and "*n*" isolated water molecules. Other structures are reported in Fig. 1.

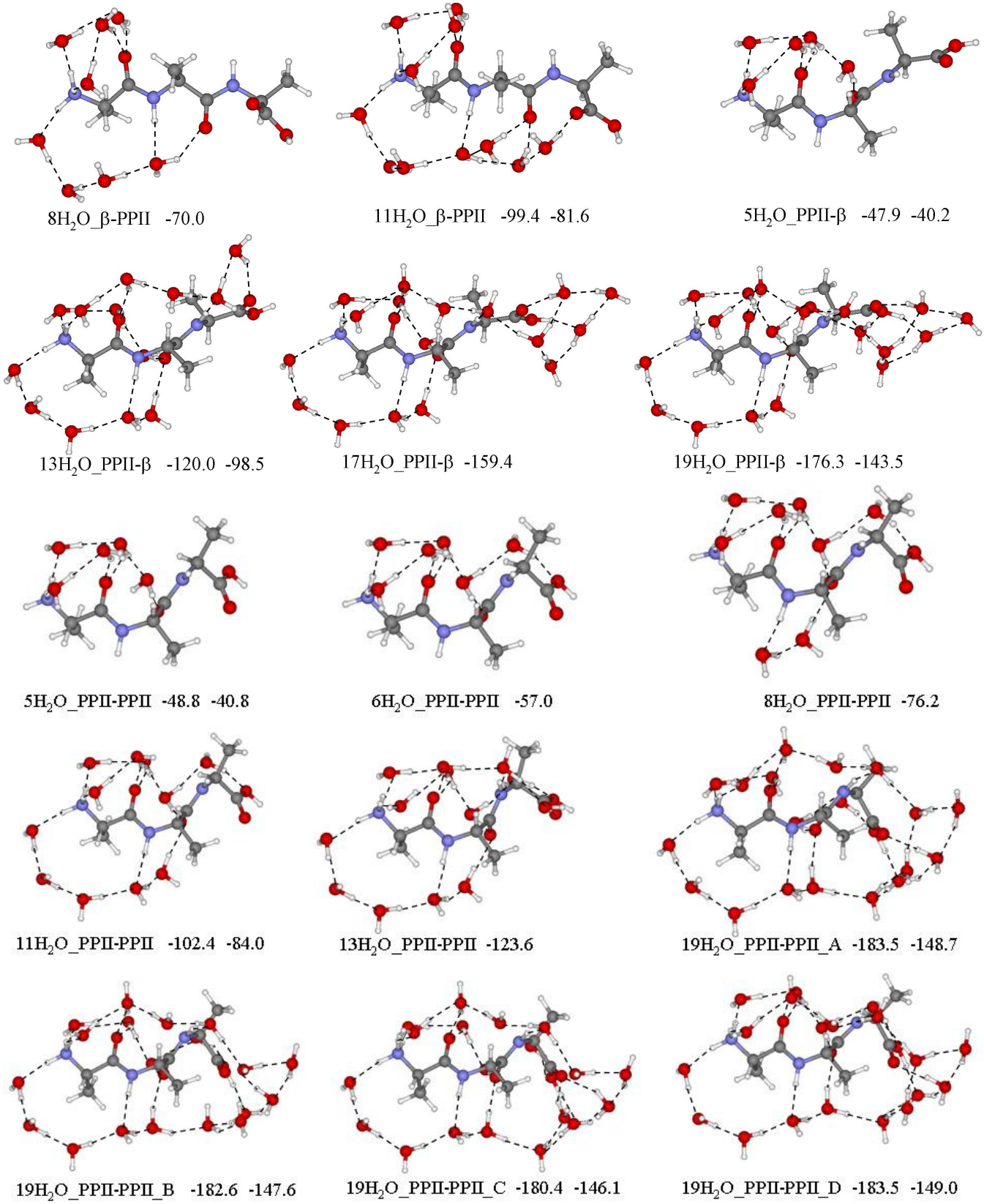

Fig. S3. Additional molecular structures of $Ala_3H^+ \cdot nH_2O$ complexes with peptidic chain in the β-PPII, PPII-β and PPII-PPII conformations. Formation energies (kcal mol^{-1}) at the 6-31+G* (left value) and aug-cc-pVTZ (right value) have been computed relative to the isolated Ala_3H^+ (in the β-β conformation) and "*n*" isolated water molecules.

Table S1. Dihedral angles [deg] for the bare Ala_3H^+ peptide and $Ala_3H^+ \cdot nH_2O$ hydrated peptide minima.

	ψ_N	ϕ_N	ψ_C	ϕ_C
C11_A	-143.5	-75.6	88.9	54.2
C11_B	141.6	70.5	-81.0	-63.3
β-β	156.7	-162.5	163.4	-159.3
$2H_2O$_β-β	151.0	-156.3	163.4	-159.5
$4H_2O$_β-β_A	141.1	-159.4	162.1	-159.7
$4H_2O$_β-β_B	138.9	-159.5	161.8	-159.9
$4H_2O$_β-β_C	139.4	-159.5	161.5	-160.2
$4H_2O$_β-β_D	140.4	-158.9	161.8	-159.9
$9H_2O$_β-β	152.8	-146.7	149.2	-157.4
$10H_2O$_β-β_A	140.5	-112.3	132.1	-158.2
$10H_2O$_β-β_C	142.5	-114.9	134.4	-157.8
$10H_2O$_β-β_D	144.2	-126.4	138.3	-157.0
$13H_2O$_β-β	156.4	-145.0	158.0	-142.0
$14H_2O$_β-β	154.1	-141.4	157.1	-149.7
$18H_2O$_β-β_A	154.0	-140.4	155.7	-151.9
$18H_2O$_β-β_B	157.7	-148.9	156.6	-149.4
$18H_2O$_β-β_C	156.0	-130.9	146.5	-148.5
$22H_2O$_β-β_A	155.1	-145.4	151.6	-146.6
$22H_2O$_β-β_B	157.3	-147.5	152.9	-144.9
$22H_2O$_β-β_C	154.7	-121.9	142.4	-142.5
$24H_2O$_β-β_A	153.5	-146.9	149.1	-146.6
$27H_2O$_β-β_A	153.0	-149.5	149.9	-148.0
$31H_2O$_β-β_A	146.1	-133.1	131.8	-137.6
$37H_2O$_β-β_A	142.1	-132.0	130.0	-138.4
β-PPII	158.8	-160.6	160.5	-64.0
$2H_2O$_β-PPII	151.5	-156.5	160.0	-63.0
$4H_2O$_β-PPII	142.6	-156.7	157.7	-63.9
$10H_2O$_β-PPII	143.9	-121.2	152.2	-60.4
$14H_2O$_β-PPII_A	148.6	-129.9	155.5	-60.3
$14H_2O$_β-PPII_B	156.8	-141.8	160.0	-60.3
$18H_2O$_β-PPII_A	155.3	-133.0	160.1	-58.9
$18H_2O$_β-PPII_A’	158.2	-140.0	161.1	-57.2
$18H_2O$_β-PPII_B	158.9	-138.3	161.6	-59.7
$18H_2O$_β-PPII_C	150.8	-114.1	150.2	-57.7
$18H_2O$_β-PPII_D	152.2	-106.4	143.7	-58.1
$22H_2O$_β-PPII_A	154.5	-127.3	155.0	-57.9
$22H_2O$_β-PPII_B	160.0	-143.8	158.4	-59.4
$22H_2O$_β-PPII_C	152.6	-100.5	141.4	-56.5
$22H_2O$_β-PPII_D	156.8	-114.1	139.3	-58.2

Table S1 continue

PPII-β	156.9	-57.4	148.2	-158.5
2H_2O_PPII-β	150.0	-54.8	144.0	-161.4
4H_2O_PPII-β_A	148.3	-58.7	157.1	-157.9
4H_2O_PPII-β_B	141.5	-56.2	150.5	-158.9
10H_2O_PPII-β_A	149.3	-55.6	148.6	-147.0
10H_2O_PPII-β_B	145.8	-52.7	138.1	-151.1
14H_2O_PPII-β_A	150.8	-58.7	154.8	-153.7
14H_2O_PPII-β_B	150.8	-58.9	155.8	-152.4
18H_2O_PPII-β_A	151.8	-59.8	156.4	-150.0
18H_2O_PPII-β_B	151.6	-59.5	154.5	-149.0
22H_2O_PPII-β_A	153.1	-57.2	159.4	-150.7
22H_2O_PPII-β_B	152.7	-56.8	157.7	-144.4
PPII-PPII	158.4	-59.3	151.6	-62.4
2H_2O_PPII-PPII	152.5	-57.0	147.2	-64.4
4H_2O_PPII-PPII	162.5	-54.5	143.3	-64.7
10H_2O_PPII-PPII_A	154.0	-53.3	139.7	-63.2
10H_2O_PPII-PPII_B	163.5	-54.2	146.2	-61.6
14H_2O_PPII-PPII_A	154.4	-57.4	143.1	-60.1
14H_2O_PPII-PPII_B	154.5	-57.3	145.2	-58.0
18H_2O_PPII-PPII_A	155.2	-56.5	148.6	-55.4
18H_2O_PPII-PPII_B	157.9	-56.9	148.4	-55.0
22H_2O_PPII-PPII_A	159.0	-55.6	149.0	-53.2
22H_2O_PPII-PPII_B	159.7	-55.7	148.1	-53.8
22H_2O_PPII-PPII_C	158.1	-56.2	149.0	-52.1
Additional structures				
8H_2O_β-PPII	143.5	-157.4	150.6	-63.6
11H_2O_β-PPII	132.9	-113.9	153.0	-62.9
5H_2O_PPII-β	152.8	-58.6	163.7	-152.9
13H_2O_PPII-β	150.4	-57.3	156.9	-152.4
17H_2O_PPII-β	151.7	-58.1	157.1	-147.1
19H_2O_PPII-β	151.5	-58.0	154.9	-143.2
5H_2O_PPII-PPII	154.9	-53.6	143.2	-64.9
6H_2O_PPII-PPII	154.2	-56.3	141.3	-65.0
8H_2O_PPII-PPII	155.6	-56.5	139.6	-64.0
11H_2O_PPII-PPII	154.9	-55.3	140.4	-65.5
13H_2O_PPII-PPII	154.4	-59.8	128.3	-65.6
19H_2O_PPII-PPII_A	157.1	-56.7	151.8	-56.7
19H_2O_PPII-PPII_B	157.0	-56.2	143.6	-56.8
19H_2O_PPII-PPII_C	157.2	-57.3	146.2	-59.5
19H_2O_PPII-PPII_D	157.0	-58.1	153.0	-57.0

Table S2. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **C11_A** conformation.

N	2.263236	-2.103324	0.642929
C	2.750633	-0.826978	0.025493
C	1.826378	0.244119	0.621644
N	1.462410	1.264046	-0.170282
C	0.372089	2.168594	0.211705
C	-0.903499	1.351810	-0.045774
N	-1.323688	0.596897	0.999783
C	-2.192029	-0.543137	0.759532
C	-1.528290	-1.455000	-0.275289
O	-2.396940	-2.189548	-0.957398
O	1.465082	0.121688	1.792594
O	-1.426182	1.305499	-1.154149
C	-3.631650	-0.157074	0.426458
O	-0.323561	-1.543585	-0.426014
C	2.779393	-0.953843	-1.487669
C	0.422767	3.442300	-0.610203
H	1.335665	-2.342367	0.257265
H	2.145181	-1.969641	1.655450
H	2.906437	-2.883590	0.481972
H	3.754452	-0.656823	0.424313
H	1.696528	1.230265	-1.156408
H	0.482330	2.383422	1.277615
H	-0.697888	0.541513	1.796935
H	-2.194239	-1.130439	1.685370
H	-1.908986	-2.789214	-1.553711
H	-3.684297	0.369454	-0.527656
H	-4.009665	0.495615	1.216798
H	-4.259496	-1.049021	0.379100
H	3.248382	-0.070515	-1.927887
H	1.771851	-1.075043	-1.895842
H	3.386017	-1.814308	-1.781902
H	-0.394045	4.105635	-0.315555
H	0.313751	3.218172	-1.674901
H	1.372379	3.956374	-0.443620

Table S3. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **C11_B** conformation.

N	-1.841122	-2.284632	0.242659
C	-2.478077	-1.089320	-0.395207
C	-1.670840	0.098695	0.133533
N	-1.432120	1.088170	-0.739680
C	-0.487791	2.193097	-0.502770
C	0.913922	1.567738	-0.566073
N	1.343162	0.936454	0.556917
C	2.397764	-0.046211	0.441364
C	1.918979	-1.213487	-0.419551
O	2.929765	-1.955098	-0.858309
O	-1.302884	0.078626	1.308526
O	1.565149	1.577459	-1.605541
C	2.807110	-0.562762	1.821748
O	0.752975	-1.467846	-0.652838
C	-3.941652	-0.991556	0.011794
C	-0.800209	3.017336	0.741303

H	-1.800137	-2.144774	1.260696
H	-0.868011	-2.390433	-0.087108
H	-2.358637	-3.147174	0.047514
H	-2.367637	-1.211011	-1.474159
H	-1.704095	0.935380	-1.703966
H	-0.561743	2.833701	-1.382766
H	0.649625	0.773017	1.283265
H	3.257576	0.405810	-0.059993
H	2.580024	-2.715925	-1.360114
H	1.958955	-1.038843	2.324510
H	3.154372	0.274190	2.431793
H	3.614382	-1.292160	1.730347
H	-4.388424	-0.114909	-0.462638
H	-4.032717	-0.887124	1.096836
H	-4.491954	-1.877432	-0.314651
H	-0.105582	3.860162	0.792156
H	-0.723504	2.440475	1.663317
H	-1.817205	3.410809	0.664142

Table S4. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **β-β** conformation.

N	5.289580	-0.256222	0.099406
C	4.058070	0.580797	0.266966
C	2.894606	-0.333064	-0.134764
N	1.700327	-0.015599	0.366785
C	0.492500	-0.731890	-0.006129
C	-0.683608	0.158884	0.394478
N	-1.864361	-0.148891	-0.166085
C	-3.079253	0.557029	0.191485
C	-4.252983	-0.323637	-0.189450
O	-5.408556	0.189753	0.233494
O	3.099858	-1.257226	-0.923005
O	-0.542532	1.066427	1.214384
C	-3.182638	1.924806	-0.495467
O	-4.162508	-1.352339	-0.821118
C	4.125329	1.796366	-0.651627
C	0.400944	-2.089333	0.701532
H	6.136761	0.310413	-0.009937
H	5.166012	-0.848554	-0.736720
H	5.438804	-0.886083	0.895078
H	4.003248	0.876096	1.315878
H	1.580210	0.750047	1.026030
H	0.499269	-0.877823	-1.091717
H	-1.943361	-0.907897	-0.835778
H	-3.095765	0.695752	1.278353
H	-6.138398	-0.389919	-0.054055
H	-3.215808	1.803661	-1.582171
H	-2.305480	2.518833	-0.230247
H	-4.080957	2.451268	-0.165285
H	3.215931	2.388948	-0.530848
H	4.203386	1.487621	-1.698281
H	4.979930	2.426621	-0.393629
H	-0.491313	-2.634416	0.381897
H	0.359447	-1.945887	1.785383
H	1.281382	-2.686360	0.453136

Table S5. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 2H_2O$ peptide-water complex in the **$2H_2O$_β–β** conformation.

N	-4.641050	-0.858722	-0.087200
C	-3.320212	-1.511467	0.142041
C	-2.275289	-0.488618	-0.302215
N	-1.095957	-0.543597	0.312382
C	0.009361	0.325227	-0.052911
C	1.290696	-0.381491	0.391069
N	2.427569	0.080543	-0.153540
C	3.723425	-0.439234	0.238353
C	4.765880	0.593208	-0.143757
O	5.976470	0.257657	0.302233
O	-2.547937	0.298139	-1.217000
O	1.259898	-1.281696	1.230143
C	4.031980	-1.789972	-0.420318
O	4.538031	1.587733	-0.795441
C	-3.209899	-2.797827	-0.669618
C	-0.111305	1.692012	0.632562
H	-5.419238	-1.489641	0.123626
H	-4.715433	-0.559356	-1.066673
H	-4.712038	0.005466	0.507664
H	-3.245670	-1.708608	1.213874
H	-0.910403	-1.215431	1.053736
H	0.001050	0.447211	-1.140832
H	2.412822	0.830192	-0.838159
H	3.738887	-0.554786	1.327942
H	6.619318	0.933628	0.016833
H	4.071222	-1.682698	-1.508216
H	3.241151	-2.497205	-0.161140
H	4.987635	-2.179600	-0.062614
H	-2.243149	-3.270029	-0.481207
H	-3.293561	-2.587212	-1.740103
H	-3.995828	-3.499346	-0.379041
H	0.682121	2.366395	0.299910
H	-0.046783	1.575290	1.718619
H	-1.076808	2.141515	0.385622
O	-3.464226	2.880698	-0.801408
H	-2.884509	3.655174	-0.837315
H	-2.966869	2.134235	-1.186649
O	-4.358047	1.462757	1.351378
H	-3.736200	1.434024	2.093535
H	-4.002983	2.119887	0.708625

Table S6. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_β–β_A** conformation.

N	4.036688	-0.628991	-1.122905
C	2.738374	-1.353417	-1.032575
C	1.761383	-0.424853	-0.320372
N	0.513122	-0.415889	-0.771330
C	-0.547053	0.346309	-0.131444
C	-1.865700	-0.283077	-0.582445
N	-2.937791	0.016561	0.167513
C	-4.268084	-0.431387	-0.196365

C	-5.263731	0.458573	0.521274
O	-6.515497	0.197826	0.144212
O	2.142789	0.236315	0.660110
O	-1.921135	-0.978682	-1.596564
C	-4.503776	-1.904884	0.159078
O	-4.967484	1.286014	1.353810
C	2.907284	-2.647277	-0.242650
C	-0.497226	1.824541	-0.536237
H	4.714670	-1.170178	-1.666303
H	4.448880	-0.464088	-0.169717
H	3.886326	0.290563	-1.593726
H	2.406669	-1.555360	-2.053689
H	0.237366	-0.966426	-1.581932
H	-0.433124	0.249031	0.953163
H	-2.850157	0.598458	0.994960
H	-4.401079	-0.292772	-1.275337
H	-7.124951	0.773376	0.643055
H	-4.426786	-2.052419	1.240260
H	-3.744275	-2.512358	-0.337769
H	-5.491952	-2.225510	-0.178123
H	1.950872	-3.172189	-0.185757
H	3.253892	-2.431828	0.772636
H	3.631004	-3.301913	-0.735103
H	-1.293954	2.385645	-0.040354
H	-0.618020	1.919794	-1.619782
H	0.463054	2.257150	-0.244195
O	3.134648	1.789343	-2.140781
H	2.957780	2.353384	-1.352782
H	2.338930	1.816944	-2.692209
O	2.737852	2.939662	0.312805
H	2.612203	2.065661	0.729344
H	3.540609	3.315353	0.703709
O	5.217835	-0.282084	1.369355
H	5.956256	-0.880860	1.553361
H	4.585923	-0.372969	2.119917
O	3.158291	-0.373051	3.176953
H	2.927046	-1.220480	3.584577
H	2.521478	-0.237682	2.450056

Table S7. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_β-β_B** conformation.

N	-3.365194	0.943988	0.320980
C	-2.249435	0.489533	-0.556876
C	-0.957549	1.014789	0.059312
N	0.083338	0.191718	0.063001
C	1.398642	0.594914	0.534798
C	2.397870	-0.386404	-0.079181
N	3.677252	0.018192	-0.087357
C	4.744236	-0.845439	-0.554892
C	6.045017	-0.317402	0.017451
O	7.063796	-1.136872	-0.242298
O	-0.910594	2.182869	0.481953
O	2.033068	-1.487867	-0.490598
C	4.815252	-0.907974	-2.086174
O	6.155900	0.731388	0.611973
C	-2.426615	1.051257	-1.964045
C	1.480964	0.551843	2.065602
H	-4.254851	0.548171	0.005231
H	-3.459412	1.991630	0.308176

H	-3.186271	0.633584	1.302155
H	-2.263554	-0.602710	-0.561507
H	0.010823	-0.761390	-0.287400
H	1.595488	1.610386	0.175780
H	3.936929	0.935668	0.261544
H	4.578236	-1.853230	-0.157852
H	7.884142	-0.748827	0.115519
H	5.032486	0.081901	-2.498085
H	3.851791	-1.249384	-2.470663
H	5.592113	-1.607221	-2.403395
H	-1.617654	0.698124	-2.607675
H	-2.410281	2.145538	-1.943082
H	-3.376602	0.715109	-2.387738
H	2.473115	0.859010	2.407303
H	1.283312	-0.463892	2.422049
H	0.742257	1.232400	2.496738
O	-2.381729	0.233340	2.811599
H	-1.891660	1.029747	3.121253
H	-1.783046	-0.522110	2.903433
O	-1.132134	2.637310	3.217638
H	-1.038804	2.783837	2.256504
H	-1.733537	3.324729	3.540364
O	-3.685420	3.703850	0.223031
H	-4.493321	3.993057	-0.225984
H	-2.933487	4.123017	-0.256271
O	-1.381218	4.453692	-1.045134
H	-0.952719	5.300554	-0.852755
H	-0.889324	3.774964	-0.545410

Table S8. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_β-β_C** conformation.

N	3.374853	-0.919292	0.240627
C	2.248499	-0.403171	-0.588154
C	0.965818	-0.988307	-0.007497
N	-0.083288	-0.178031	0.061788
C	-1.389699	-0.627763	0.514477
C	-2.406345	0.381129	-0.021002
N	-3.679974	-0.039939	-0.052027
C	-4.762406	0.841123	-0.445892
C	-6.052037	0.247862	0.086586
O	-7.084583	1.069420	-0.101819
O	0.932679	-2.184913	0.326430
O	-2.058822	1.514982	-0.351070
C	-4.841233	1.023853	-1.967159
O	-6.143615	-0.847552	0.593941
C	2.417144	-0.844314	-2.038812
C	-1.457055	-0.687244	2.045216
H	4.257170	-0.482822	-0.040142
H	3.484301	-1.959766	0.130292
H	3.196506	-0.703176	1.246553
H	2.253699	0.685747	-0.502040
H	-0.021568	0.799556	-0.215466
H	-1.577803	-1.619229	0.090051
H	-3.924803	-0.984788	0.227681
H	-4.610821	1.816819	0.029421
H	-7.897222	0.640628	0.225796
H	-5.043152	0.066138	-2.455768
H	-3.885363	1.411428	-2.325928
H	-5.631293	1.732443	-2.225573

H	1.603138	-0.439722	-2.644703
H	2.402316	-1.936442	-2.109988
H	3.363576	-0.472129	-2.439726
H	-2.433033	-1.051409	2.376742
H	-1.287169	0.307919	2.467405
H	-0.688371	-1.368293	2.419335
O	2.398989	-0.456528	2.796627
H	1.964659	-1.302102	3.050846
H	1.761461	0.253775	2.958877
O	1.446239	-3.004508	2.940377
H	1.120305	-2.997909	2.020510
H	0.744922	-3.391641	3.484007
O	3.739073	-3.650053	-0.126858
H	4.535265	-3.873118	-0.631134
H	2.978285	-4.035557	-0.620466
O	1.410245	-4.315925	-1.394353
H	1.000709	-5.183065	-1.259125
H	0.921019	-3.688186	-0.829616

Table S9. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_β-β_D** conformation.

N	-3.350764	0.907924	0.279577
C	-2.227556	0.431276	-0.575253
C	-0.939464	0.954594	0.050144
N	0.101050	0.130331	0.047382
C	1.415811	0.527067	0.524512
C	2.417675	-0.438818	-0.109274
N	3.696039	-0.030113	-0.111043
C	4.765115	-0.882925	-0.593484
C	6.064903	-0.360655	-0.013679
O	7.088008	-1.165907	-0.299656
O	-0.893818	2.117412	0.485529
O	2.054901	-1.532911	-0.541205
C	4.832864	-0.920893	-2.125564
O	6.171446	0.672431	0.608307
C	-2.378837	0.969459	-1.994476
C	1.500199	0.452549	2.053652
H	-4.238194	0.507560	-0.036656
H	-3.440597	1.954841	0.239656
H	-3.186783	0.620128	1.270993
H	-2.248988	-0.660823	-0.562802
H	0.027078	-0.818068	-0.314911
H	1.609411	1.550114	0.186131
H	3.954095	0.880220	0.257369
H	4.602856	-1.897423	-0.212041
H	7.906619	-0.784808	0.069357
H	5.046708	0.076136	-2.521640
H	3.868649	-1.257894	-2.512094
H	5.610050	-1.613467	-2.456216
H	-1.555049	0.609907	-2.615462
H	-2.367691	2.063737	-1.989391
H	-3.318412	0.624757	-2.434024
H	2.485824	0.771932	2.402262
H	1.318295	-0.572621	2.390101
H	0.746433	1.109550	2.494788
O	-2.454274	0.246338	2.821843
H	-2.001742	1.061598	3.137189
H	-1.824148	-0.482168	2.921322
O	-1.402104	2.738888	3.158755

H	-1.058863	2.787080	2.246670
H	-0.693094	3.044770	3.742670
O	-3.681191	3.661476	0.096227
H	-4.432789	3.943728	-0.445110
H	-2.886753	4.122599	-0.260155
O	-1.237545	4.613523	-0.697773
H	-1.048230	4.722730	-1.641197
H	-0.821604	3.772220	-0.430721

Table S10. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 9H_2O$ peptide-water complex in the **$9H_2O$_β-β** conformation.

N	3.833805	-0.589659	-0.002948
C	2.607754	0.203285	-0.266866
C	1.401129	-0.672029	0.046418
N	0.297828	-0.025113	0.412271
C	-0.932917	-0.726882	0.736134
C	-2.110780	0.138053	0.308431
N	-3.212877	-0.505993	-0.071033
C	-4.454395	0.188775	-0.367641
C	-5.581195	-0.814940	-0.206032
O	-6.773511	-0.232026	-0.313668
O	1.474628	-1.907030	-0.083292
O	-2.049781	1.378786	0.370175
C	-4.465941	0.788617	-1.778588
O	-5.417537	-2.000919	-0.029096
C	2.583745	0.667698	-1.720406
C	-1.028168	-1.016568	2.240292
H	4.641690	0.059862	-0.081785
H	3.941455	-1.372547	-0.684314
H	3.790646	-0.964115	0.963804
H	2.626901	1.062911	0.407867
H	0.337892	0.997158	0.497860
H	-0.934357	-1.665246	0.174518
H	-3.234047	-1.521768	-0.102074
H	-4.591324	0.985283	0.371575
H	-7.469956	-0.910666	-0.234783
H	-4.341046	0.002672	-2.528593
H	-3.646745	1.503964	-1.874869
H	-5.407921	1.311279	-1.957991
H	1.717375	1.311719	-1.889816
H	2.525847	-0.190600	-2.397853
H	3.488674	1.239568	-1.944425
H	-1.958288	-1.542731	2.472953
H	-0.996833	-0.081608	2.807556
H	-0.186777	-1.645675	2.542363
O	3.118416	-1.287377	2.627440
H	2.520338	-2.068287	2.643837
H	2.702594	-0.607876	3.177411
O	1.437541	-3.435269	2.230978
H	1.336444	-3.172897	1.295164
H	1.883001	-4.295255	2.225453
O	4.174685	-2.575020	-1.978997
H	4.632221	-2.237630	-2.762762
H	3.301848	-2.909185	-2.288896
O	1.593246	-3.384534	-2.443857
H	1.128511	-3.013330	-3.207991
H	1.285006	-2.879800	-1.666554

O	0.951728	2.826841	0.281768
H	0.352606	3.061566	-0.464884
H	0.476094	3.147977	1.080189
O	5.449318	1.674578	-0.217558
H	6.074598	1.860277	0.497747
H	4.796724	2.414531	-0.211652
O	3.499343	3.583165	-0.369481
H	2.578337	3.393315	-0.072874
H	3.720740	4.459500	-0.022680
O	-1.065087	3.162594	-1.549014
H	-0.930332	2.786338	-2.431439
H	-1.589695	2.506212	-1.051005
O	-0.983276	3.281945	2.170138
H	-1.506035	2.578695	1.738655
H	-0.890556	3.019812	3.097536

Table S11. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}10H_2O$ peptide-water complex in the **$10H_2O$_β-β_A** conformation.

N	-3.276502	1.408519	0.126979
C	-2.242467	0.445193	-0.329764
C	-0.925059	0.832790	0.331107
N	-0.164025	-0.175607	0.748031
C	1.156127	0.004306	1.331529
C	2.196890	-0.567924	0.371619
N	3.255569	0.194976	0.104173
C	4.348653	-0.268184	-0.733105
C	5.566942	0.569030	-0.392787
O	6.659430	0.110928	-1.001333
O	-0.618199	2.034494	0.425550
O	2.063901	-1.708882	-0.105146
C	4.018650	-0.159724	-2.226851
O	5.545447	1.547772	0.318937
C	-2.109518	0.502471	-1.848583
C	1.243426	-0.708826	2.680796
H	-4.199194	1.134454	-0.279032
H	-3.060999	2.376909	-0.198104
H	-3.319277	1.384548	1.161034
H	-2.556392	-0.550894	-0.002906
H	-0.525909	-1.135249	0.656587
H	1.315441	1.076149	1.463991
H	3.347415	1.118216	0.518236
H	4.562140	-1.311946	-0.479545
H	7.414261	0.689111	-0.782817
H	3.825083	0.881721	-2.499282
H	3.128214	-0.754411	-2.442132
H	4.849307	-0.539760	-2.825490
H	-1.376492	-0.234951	-2.185170
H	-1.782444	1.497778	-2.166194
H	-3.071665	0.280438	-2.318071
H	2.248357	-0.608926	3.100551
H	1.012433	-1.772754	2.568343
H	0.526148	-0.256148	3.369196
O	-2.681480	0.848464	2.824300
H	-1.845481	1.288829	3.098527
H	-2.581190	-0.088540	3.047760
O	-0.327339	2.245994	3.172378
H	-0.302648	2.540988	2.241586

H	-0.405934	3.047714	3.710429
O	-2.912422	3.971429	-0.999744
H	-3.450316	4.070528	-1.798713
H	-1.979102	4.122851	-1.273311
O	-0.201022	4.081363	-1.395963
H	0.165724	3.839404	-2.258738
H	-0.104231	3.293603	-0.825935
O	-1.227145	-2.801652	0.272715
H	-0.926782	-2.840432	-0.663869
H	-0.584366	-3.382719	0.738934
O	-5.493098	0.517227	-1.293612
H	-6.392881	0.664743	-0.967988
H	-5.408640	-0.455467	-1.453392
O	-5.076978	-2.130744	-1.685427
H	-4.504370	-2.333437	-2.439164
H	-4.642275	-2.523978	-0.890676
O	-3.918305	-3.164770	0.559194
H	-2.937076	-3.085663	0.514091
H	-4.191494	-2.696220	1.361362
O	0.287768	-2.762729	-1.998898
H	0.182031	-2.350459	-2.868099
H	1.002090	-2.277645	-1.542259
O	1.065253	-4.079195	1.033440
H	1.605986	-3.316122	0.748055
H	1.410876	-4.359856	1.893004

Table S12. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_β-β_C** conformation.

N	3.308713	-1.348573	0.060408
C	2.252802	-0.380853	-0.331342
C	0.947176	-0.831402	0.312652
N	0.150554	0.138099	0.754153
C	-1.166578	-0.110374	1.318297
C	-2.216958	0.510626	0.399869
N	-3.283193	-0.232667	0.108916
C	-4.384124	0.281158	-0.687888
C	-5.606288	-0.558586	-0.368965
O	-6.700964	-0.065167	-0.944891
O	0.675629	-2.044583	0.361426
O	-2.084127	1.673731	-0.018941
C	-4.077213	0.244539	-2.189912
O	-5.586434	-1.566014	0.301637
C	2.109196	-0.354600	-1.850977
C	-1.276730	0.502735	2.715180
H	4.220851	-1.038652	-0.343171
H	3.100875	-2.302252	-0.310717
H	3.365927	-1.379467	1.094185
H	2.549227	0.601191	0.049448
H	0.481657	1.111682	0.697792
H	-1.302963	-1.193110	1.364520
H	-3.377830	-1.173751	0.479852
H	-4.582510	1.313435	-0.380024
H	-7.459191	-0.644152	-0.740983
H	-3.907614	-0.784197	-2.520395
H	-3.178453	0.833152	-2.386758
H	-4.908629	0.671619	-2.754830
H	1.365423	0.391033	-2.142733
H	1.791623	-1.334293	-2.222513
H	3.065785	-0.093308	-2.311725

H	-2.278820	0.347040	3.123813
H	-1.074837	1.577664	2.677178
H	-0.546863	0.031986	3.378590
O	2.754885	-1.039075	2.800276
H	2.039088	-1.664828	3.052926
H	2.476130	-0.162569	3.102896
O	0.822590	-2.983467	2.984881
H	0.546821	-2.848126	2.057200
H	0.019271	-2.916989	3.521533
O	2.923002	-3.849982	-1.192235
H	3.499919	-3.934805	-1.965320
H	2.000289	-3.924573	-1.526860
O	0.261249	-3.691591	-1.832551
H	-0.291126	-4.478677	-1.718716
H	0.123975	-3.143080	-1.035913
O	1.161897	2.803654	0.384789
H	0.848542	2.890286	-0.544432
H	0.518298	3.347751	0.892282
O	5.501932	-0.337691	-1.319535
H	6.402992	-0.477434	-0.994063
H	5.394797	0.638687	-1.437302
O	5.031324	2.315367	-1.604010
H	4.459783	2.535965	-2.353526
H	4.585717	2.673791	-0.798755
O	3.840564	3.258609	0.663168
H	2.863113	3.140402	0.619252
H	4.135250	2.786430	1.455452
O	-0.364856	2.860289	-1.882142
H	-0.247558	2.487958	-2.767801
H	-1.064678	2.336077	-1.447118
O	-1.136919	3.991280	1.260940
H	-1.669150	3.239947	0.932554
H	-1.483528	4.217810	2.135861

Table S13. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_β-β_D** conformation.

N	3.424261	-1.184993	0.055372
C	2.312234	-0.290499	-0.353292
C	1.020595	-0.867500	0.215265
N	0.114022	0.018058	0.619215
C	-1.188870	-0.367053	1.136820
C	-2.265920	0.370744	0.346954
N	-3.347934	-0.327864	0.007036
C	-4.484789	0.287789	-0.656141
C	-5.696293	-0.582651	-0.381482
O	-6.816302	-0.013466	-0.823406
O	0.860154	-2.100973	0.239950
O	-2.143942	1.579385	0.082928
C	-4.259401	0.443202	-2.164989
O	-5.649053	-1.669989	0.147734
C	2.237446	-0.219136	-1.876256
C	-1.312432	-0.013089	2.621763
H	4.326405	-0.790794	-0.293915
H	3.310445	-2.138307	-0.354666
H	3.439032	-1.246351	1.088772
H	2.508530	0.697504	0.073955
H	0.365994	1.016413	0.616939
H	-1.290835	-1.445420	0.993422
H	-3.427866	-1.311068	0.250292

H	-4.655438	1.272003	-0.207401
H	-7.568364	-0.611512	-0.654622
H	-4.119653	-0.535683	-2.632274
H	-3.365371	1.047704	-2.333345
H	-5.114339	0.942877	-2.625488
H	1.453814	0.478757	-2.180748
H	2.015500	-1.206153	-2.295044
H	3.191695	0.129145	-2.280962
H	-2.304858	-0.276503	2.997981
H	-1.148001	1.058099	2.773591
H	-0.561262	-0.565871	3.191815
O	2.729141	-0.969799	2.772761
H	2.058731	-1.651865	3.000058
H	2.375295	-0.119819	3.072692
O	0.971415	-3.078861	2.843088
H	0.711617	-2.951220	1.909855
H	0.152181	-3.199302	3.344709
O	3.313025	-3.663210	-1.293316
H	3.845932	-3.625913	-2.100979
H	2.395903	-3.880633	-1.577554
O	0.628597	-4.030680	-1.738977
H	0.256495	-3.762228	-2.591633
H	0.435979	-3.304321	-1.115190
O	1.010373	2.751165	0.466126
H	0.663202	2.927319	-0.438597
H	0.367025	3.214339	1.049906
O	5.614543	0.011392	-1.175223
H	6.486397	-0.028454	-0.756028
H	5.421121	0.969806	-1.326005
O	4.909825	2.599920	-1.542884
H	4.325642	2.761433	-2.297557
H	4.426736	2.914984	-0.741035
O	3.646413	3.401333	0.737454
H	2.680674	3.207348	0.693245
H	3.983066	2.922053	1.508477
O	-0.582218	3.015619	-1.744118
H	-0.422659	2.655602	-2.628445
H	-1.242633	2.432400	-1.322497
O	-1.262581	3.787528	1.574694
H	-1.784352	3.044848	1.213369
H	-1.534767	3.896612	2.497197

Table S14. M062 X/6-31+G*/PC M optimized Cartesian coordinates of the $Ala_3H^+ \cdot 13H_2O$ peptide-water complex in the **$13H_2O$_β-β** conformation.

N	-3.652063	1.094073	0.128043
C	-2.691736	-0.011287	-0.116318
C	-1.293138	0.552922	0.087745
N	-0.340349	-0.314127	0.393557
C	1.038834	0.101189	0.587055
C	1.958030	-0.999184	0.069610
N	3.190901	-0.617683	-0.245410
C	4.184423	-1.569334	-0.702108
C	5.532173	-1.190268	-0.115498
O	6.483042	-2.048279	-0.499748
O	-1.088601	1.771222	-0.096475
O	1.566911	-2.181706	0.002424
C	4.233115	-1.650138	-2.232346
O	5.738406	-0.240953	0.603427
C	-2.851607	-0.550331	-1.535612

C	1.328597	0.382170	2.066945
H	-4.628312	0.765396	-0.045043
H	-3.466318	1.901313	-0.505919
H	-3.545612	1.407964	1.106705
H	-2.887669	-0.793532	0.620592
H	-0.605564	-1.290123	0.569604
H	1.202148	1.012232	0.004445
H	3.426325	0.385613	-0.204945
H	3.917008	-2.551286	-0.297202
H	7.331023	-1.776544	-0.101955
H	4.506048	-0.680079	-2.658220
H	3.244679	-1.932561	-2.603651
H	4.959864	-2.398684	-2.554481
H	-2.124690	-1.347403	-1.716147
H	-2.689081	0.246792	-2.269183
H	-3.857858	-0.956983	-1.670337
H	2.362327	0.713571	2.199914
H	1.166262	-0.520258	2.665356
H	0.657164	1.167233	2.426149
O	-2.399153	1.245848	2.630859
H	-1.822555	2.025907	2.758518
H	-2.736288	1.004553	3.505759
O	-0.657891	3.361791	2.085583
H	-0.812484	2.981731	1.193212
H	-1.131274	4.208321	2.105170
O	-3.344956	3.283508	-1.666430
H	-3.964302	3.293639	-2.410514
H	-2.449988	3.389419	-2.050555
O	-0.608041	3.242328	-2.320385
H	-0.398127	2.829356	-3.172026
H	-0.626168	2.519585	-1.658727
O	-1.614923	-2.931110	0.615854
H	-1.335321	-3.183263	-0.293559
H	-0.984302	-3.398721	1.210939
O	-6.139121	0.117342	-0.658617
H	-6.941639	0.364100	-0.176577
H	-6.148860	-0.868543	-0.733128
O	-5.977146	-2.580841	-0.835288
H	-5.725351	-2.950664	-1.693502
H	-5.319351	-2.915127	-0.178871
O	-4.254787	-3.438996	1.090535
H	-3.292800	-3.279823	0.941181
H	-4.466639	-3.031001	1.942511
O	3.442215	2.216375	-0.116571
H	3.159766	2.639052	0.716672
H	2.963315	2.748345	-0.786746
O	2.005437	3.955192	1.578728
H	1.109985	3.760468	1.933254
H	2.388418	4.632466	2.155584
O	1.854289	4.200175	-1.250292
H	0.983584	3.987823	-1.640322
H	1.703137	4.365910	-0.299553
O	0.570313	-3.921071	1.951781
H	0.708332	-3.644684	2.869531
H	1.122153	-3.327653	1.404437
O	-0.360012	-3.087556	-1.807687
H	0.475188	-2.836839	-1.364381
H	-0.175602	-3.903431	-2.295744

Table S15. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_β-β** conformation.

N	-3.750355	1.386392	0.045856
C	-2.930026	0.169900	-0.184326
C	-1.475517	0.572167	0.016087
N	-0.631949	-0.383639	0.381364
C	0.779471	-0.112512	0.594101
C	1.591895	-1.287776	0.063274
N	2.854723	-1.022277	-0.254741
C	3.747928	-2.069000	-0.712497
C	5.159360	-1.715568	-0.289059
O	6.028926	-2.660256	-0.635671
O	-1.132338	1.746046	-0.220959
O	1.097288	-2.429662	-0.016292
C	3.654470	-2.282404	-2.228642
O	5.468458	-0.694984	0.287378
C	-3.146313	-0.356389	-1.600303
C	1.079012	0.117910	2.080014
H	-4.757339	1.176719	-0.132848
H	-3.454720	2.162469	-0.584077
H	-3.613302	1.684576	1.027712
H	-3.223624	-0.575842	0.558640
H	-1.011421	-1.312229	0.601347
H	1.033445	0.788812	0.030143
H	3.192912	-0.051598	-0.186285
H	3.472981	-2.998363	-0.201640
H	6.923052	-2.394328	-0.349937
H	3.947663	-1.373946	-2.762699
H	2.620617	-2.525576	-2.485998
H	4.299531	-3.106531	-2.540385
H	-2.541787	-1.253674	-1.761505
H	-2.856721	0.400690	-2.337378
H	-4.199766	-0.610377	-1.747723
H	2.140998	0.334687	2.225271
H	0.811793	-0.766627	2.667366
H	0.488161	0.967793	2.434567
O	-2.548400	1.444620	2.572516
H	-1.891492	2.178765	2.608540
H	-2.983503	1.418281	3.436807
O	-0.830796	3.460496	1.968031
H	-0.945653	3.104037	1.067140
H	0.146471	3.493426	2.071377
O	-3.092753	3.559635	-1.698769
H	-3.669327	3.726770	-2.458235
H	-2.176778	3.567598	-2.043190
O	-0.332530	3.205489	-2.322982
H	-0.175907	2.814224	-3.196235
H	-0.476033	2.457925	-1.703912
O	-2.144393	-2.852051	0.679065
H	-1.920870	-3.130739	-0.237854
H	-1.544133	-3.381613	1.253588
O	-6.333349	0.636534	-0.692200
H	-7.092647	0.979205	-0.198935
H	-6.429003	-0.347668	-0.703696
O	-6.415503	-2.071196	-0.713424
H	-6.174533	-2.495779	-1.549091
H	-5.804674	-2.433186	-0.026797
O	-4.802381	-3.002907	1.273851
H	-3.834400	-2.979068	1.085404
H	-4.930416	-2.500914	2.091560
O	3.352230	1.774997	0.127224
H	4.310595	1.952528	0.266901

H	3.046149	2.425424	-0.537904
O	1.901459	3.629693	1.684437
H	2.396356	2.804507	1.516408
H	1.875043	4.030429	0.792610
O	2.064687	3.953837	-1.122255
H	1.235390	3.796431	-1.627460
H	2.536050	4.664459	-1.581628
O	-0.037579	-4.084501	1.932921
H	0.159452	-3.837855	2.848506
H	0.561656	-3.550368	1.374532
O	-0.964397	-3.130994	-1.772997
H	-0.106896	-2.950056	-1.338274
H	-0.852231	-3.960512	-2.259942
O	6.074525	2.023002	0.540055
H	6.225100	1.064086	0.607752
H	6.349899	2.399319	1.388778

Table S16. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-β_A** conformation.

N	-4.813835	1.371924	0.123815
C	-4.001881	0.154929	-0.132477
C	-2.543225	0.553022	0.042831
N	-1.693642	-0.405731	0.385502
C	-0.277672	-0.138481	0.570442
C	0.524874	-1.299850	-0.005043
N	1.766641	-1.011369	-0.370003
C	2.677837	-2.028189	-0.861744
C	4.095987	-1.593469	-0.530069
O	5.006391	-2.423850	-0.964545
O	-2.204968	1.729080	-0.190845
O	0.040113	-2.448000	-0.075083
C	2.500604	-2.276660	-2.363152
O	4.332658	-0.553513	0.072772
C	-4.247936	-0.358653	-1.548226
C	0.058858	0.061262	2.052682
H	-5.824867	1.166142	-0.034582
H	-4.529761	2.151517	-0.507094
H	-4.653407	1.664071	1.103690
H	-4.282750	-0.596120	0.609958
H	-2.069424	-1.336513	0.602415
H	-0.038661	0.775179	0.019195
H	2.100987	-0.040406	-0.286972
H	2.480940	-2.958918	-0.316946
H	5.953872	-2.021944	-0.843931
H	2.724165	-1.367680	-2.929795
H	1.465785	-2.568879	-2.558648
H	3.164297	-3.076923	-2.696666
H	-3.649335	-1.255987	-1.729836
H	-3.971321	0.404266	-2.284238
H	-5.304882	-0.608772	-1.676300
H	1.124518	0.276407	2.174374
H	-0.190802	-0.836027	2.628324
H	-0.522963	0.902680	2.440704
O	-3.557150	1.402440	2.626434
H	-2.872105	2.109741	2.664272
H	-3.975878	1.374850	3.498747
O	-1.761774	3.357770	2.035227

H	-1.896907	3.019807	1.129811
H	-0.783142	3.385421	2.123338
O	-4.185914	3.560270	-1.612941
H	-4.763105	3.727042	-2.372016
H	-3.269610	3.582099	-1.955675
O	-1.421108	3.248057	-2.255347
H	-1.268831	2.885089	-3.141521
H	-1.553500	2.480666	-1.658611
O	-3.202263	-2.877315	0.674574
H	-2.992108	-3.143506	-0.249309
H	-2.592066	-3.413203	1.232530
O	-7.410379	0.635906	-0.579214
H	-8.165308	0.971426	-0.074473
H	-7.508534	-0.347573	-0.608788
O	-7.494224	-2.071128	-0.649794
H	-7.270313	-2.485090	-1.495475
H	-6.872655	-2.444813	0.020661
O	-5.852943	-3.033772	1.299344
H	-4.887172	-3.008766	1.100021
H	-5.970764	-2.542271	2.124934
O	2.344068	1.768557	0.002055
H	3.313861	1.913063	0.057839
H	2.002024	2.452923	-0.609525
O	0.967717	3.549455	1.707437
H	1.465553	2.737932	1.489433
H	0.903431	3.984552	0.833887
O	0.994670	3.991951	-1.088294
H	0.150259	3.838817	-1.569488
H	1.436422	4.728538	-1.536026
O	-1.067872	-4.116690	1.870454
H	-0.849970	-3.890547	2.786539
H	-0.481899	-3.569939	1.309439
O	-2.055699	-3.126207	-1.795276
H	-1.193324	-2.949105	-1.367689
H	-1.949147	-3.952710	-2.288552
O	5.985560	0.460048	1.990226
H	5.584435	0.420678	2.871987
H	5.434901	-0.109662	1.409201
O	5.097433	2.347516	0.204698
H	5.471976	1.889091	0.989712
H	5.297431	3.290049	0.308401
O	8.372719	-0.864875	1.580175
H	9.169011	-0.314150	1.572866
H	7.642518	-0.293548	1.901056
O	7.299147	-1.335990	-0.904745
H	7.743787	-1.219924	-0.032476
H	7.075234	-0.431060	-1.232832
O	6.425584	1.069221	-1.885267
H	5.905253	1.521185	-1.184804
H	5.790887	0.852139	-2.584094

Table S17. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}18H_2O$ peptide-water complex in the **$18H_2O$_β-β_B** conformation.

N	-4.850427	1.348169	0.240995
C	-4.005599	0.164440	-0.056324
C	-2.556409	0.590000	0.132978
N	-1.673078	-0.367136	0.368349
C	-0.260520	-0.066922	0.540211

C	0.555380	-1.249162	0.031948
N	1.801401	-0.975479	-0.333007
C	2.717888	-2.019954	-0.754231
C	4.129342	-1.599529	-0.376503
O	5.043020	-2.443652	-0.779381
O	-2.248943	1.793936	-0.000799
O	0.075899	-2.400785	0.018387
C	2.596193	-2.314751	-2.252414
O	4.356027	-0.562386	0.231487
C	-4.248602	-0.307326	-1.487508
C	0.070593	0.210929	2.011925
H	-5.858905	1.115622	0.095327
H	-4.604527	2.145896	-0.383287
H	-4.687005	1.625250	1.222837
H	-4.262534	-0.620149	0.659344
H	-2.011014	-1.329519	0.493977
H	-0.028537	0.818920	-0.058625
H	2.134276	-0.001850	-0.303328
H	2.480791	-2.927705	-0.187682
H	5.991120	-2.050362	-0.645223
H	2.860148	-1.429513	-2.838881
H	1.564709	-2.594186	-2.481480
H	3.256753	-3.137552	-2.532507
H	-3.633026	-1.185022	-1.699407
H	-3.995841	0.482279	-2.203132
H	-5.299013	-0.582523	-1.617898
H	1.129175	0.462344	2.125885
H	-0.153156	-0.667922	2.625064
H	-0.531742	1.051774	2.367033
O	-3.550488	1.281505	2.723642
H	-2.914130	2.008650	2.878960
H	-3.905443	1.033860	3.589705
O	-1.677298	3.278099	2.223078
H	-1.921249	2.971700	1.323877
H	-2.034092	4.175245	2.313458
O	-4.341833	3.478037	-1.589786
H	-4.969488	3.541542	-2.324233
H	-3.448751	3.485729	-1.990354
O	-1.614925	3.123205	-2.279783
H	-1.516968	2.592091	-3.085276
H	-1.708693	2.484783	-1.540046
O	-3.142037	-2.872425	0.531275
H	-2.844157	-3.239684	-0.332323
H	-2.589109	-3.342574	1.197583
O	-7.459569	0.619394	-0.410457
H	-8.170326	0.852967	0.203975
H	-7.544527	-0.351389	-0.578029
O	-7.519179	-2.053866	-0.841104
H	-7.330164	-2.371446	-1.735500
H	-6.870635	-2.494150	-0.240156
O	-5.822752	-3.186195	0.958247
H	-4.851096	-3.095375	0.815182
H	-5.998648	-2.836157	1.843571
O	2.376241	1.857128	-0.233272
H	1.981836	2.403371	-0.943222
H	2.004340	2.281856	0.572867
O	1.019699	3.585099	1.398140
H	0.158467	3.441042	1.837550
H	0.804700	3.968357	0.524381
O	0.886793	3.945284	-1.397460
H	0.016190	3.773332	-1.820515
H	1.270106	4.708335	-1.854375
O	-1.110241	-3.948347	2.027652
H	-0.968442	-3.655982	2.939817

H	-0.488273	-3.433070	1.476683
O	-1.731538	-3.669050	-1.681668
H	-0.954825	-3.203229	-1.313939
H	-1.870569	-3.310936	-2.570365
O	6.099460	0.751565	1.877781
H	5.698070	0.832871	2.756353
H	5.536691	0.120396	1.377064
O	5.129246	2.421290	-0.197263
H	5.491069	2.054764	0.632908
H	4.183156	2.155543	-0.206759
O	8.436439	-0.680523	1.691201
H	9.250254	-0.161554	1.616347
H	7.721002	-0.046212	1.916111
O	7.349218	-1.388017	-0.733049
H	7.792523	-1.196642	0.126272
H	7.140589	-0.515697	-1.150725
O	6.527909	0.899856	-1.974677
H	5.967975	1.447102	-1.367783
H	5.946886	0.626345	-2.699482

Table S18. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}18H_2O$ peptide-water complex in the **$18H_2O$_β-β_C** conformation.

N	-4.730442	1.590058	0.005746
C	-3.963008	0.332813	-0.177701
C	-2.504801	0.646235	0.131311
N	-1.747146	-0.373950	0.505372
C	-0.337466	-0.227008	0.830120
C	0.457733	-1.290210	0.080411
N	1.675924	-0.938501	-0.310373
C	2.566433	-1.882797	-0.961383
C	3.997886	-1.528023	-0.591835
O	4.883910	-2.287087	-1.182702
O	-2.084955	1.812530	-0.024288
O	-0.015076	-2.431505	-0.094279
C	2.359646	-1.904031	-2.479020
O	4.261587	-0.611434	0.173974
C	-4.120362	-0.179142	-1.606909
C	-0.112790	-0.376240	2.338345
H	-5.741224	1.418085	-0.195820
H	-4.388492	2.334494	-0.639177
H	-4.609130	1.907038	0.981704
H	-4.347250	-0.396767	0.539917
H	-2.195896	-1.290353	0.635114
H	-0.018979	0.764838	0.498463
H	2.003558	0.025563	-0.149733
H	2.358770	-2.879412	-0.555589
H	5.841914	-1.927339	-1.028221
H	2.584016	-0.922974	-2.908961
H	1.317853	-2.153732	-2.695651
H	3.007655	-2.651811	-2.940264
H	-3.566321	-1.113584	-1.728575
H	-3.738396	0.552816	-2.326156
H	-5.175381	-0.369122	-1.821915
H	0.950435	-0.279208	2.576517
H	-0.463498	-1.354180	2.683541
H	-0.670689	0.402108	2.867196
O	-3.636756	1.531216	2.584343
H	-2.861779	2.104149	2.756232
H	-4.065332	1.380831	3.439346

O	-1.291831	3.032684	2.295131
H	-1.437741	2.787258	1.355861
H	-1.457475	3.986871	2.352588
O	-3.949121	3.574156	-1.878885
H	-4.514638	3.646007	-2.661557
H	-3.032081	3.478841	-2.209584
O	-1.245444	2.951219	-2.357255
H	-1.144931	2.346153	-3.108445
H	-1.398569	2.386142	-1.569807
O	-3.317975	-2.795370	0.613148
H	-3.130153	-3.065721	-0.313198
H	-2.739076	-3.385750	1.149589
O	-7.306398	0.914382	-0.795721
H	-8.072291	1.325122	-0.369130
H	-7.454508	-0.063041	-0.754098
O	-7.547431	-1.778125	-0.689485
H	-7.340134	-2.234287	-1.517579
H	-6.954561	-2.164724	-0.000666
O	-5.966348	-2.797270	1.281566
H	-5.004279	-2.828152	1.068429
H	-6.044720	-2.299160	2.107929
O	2.279412	1.846737	0.162867
H	1.889560	2.517447	-0.444242
H	2.015753	2.124207	1.070620
O	1.440129	2.670300	2.653449
H	0.464390	2.777396	2.637845
H	1.644609	2.121750	3.424193
O	1.255332	3.821427	-1.479358
H	0.402793	3.599564	-1.911091
H	1.854854	4.099870	-2.186467
O	-1.244355	-4.256996	1.623568
H	-0.960754	-4.280593	2.548923
H	-0.618899	-3.668217	1.155278
O	-2.051487	-3.378059	-1.740359
H	-1.226729	-2.988888	-1.387830
H	-2.119938	-3.099868	-2.664709
O	6.076120	0.453829	1.917394
H	5.723418	0.414547	2.819298
H	5.483513	-0.102480	1.365558
O	5.021863	2.389330	0.126690
H	5.420549	1.912301	0.880017
H	4.074008	2.123469	0.124872
O	8.399171	-0.912507	1.380973
H	9.201822	-0.372863	1.337188
H	7.692544	-0.331713	1.739069
O	7.205669	-1.271121	-1.071089
H	7.681484	-1.204174	-0.210538
H	6.984138	-0.347980	-1.349830
O	6.326741	1.164810	-1.923588
H	5.797474	1.605429	-1.210721
H	5.708712	0.995812	-2.649741

Table S19. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-β_A** conformation.

N	5.345514	-1.141183	-0.100067
C	4.322504	-0.067231	-0.169397
C	2.965020	-0.748755	-0.058044
N	1.960302	-0.019316	0.403658

C	0.622739	-0.572454	0.548945
C	-0.384953	0.527298	0.244335
N	-1.554436	0.131982	-0.239914
C	-2.638232	1.068115	-0.484366
C	-3.945056	0.344497	-0.214523
O	-5.021934	1.051531	-0.480672
O	2.844956	-1.926730	-0.447555
O	-0.111389	1.718128	0.490900
C	-2.574004	1.655930	-1.896747
O	-3.990116	-0.798791	0.215304
C	4.438344	0.690498	-1.488639
C	0.398174	-1.107694	1.967730
H	6.303217	-0.736498	-0.198998
H	5.199788	-1.843460	-0.856305
H	5.250076	-1.615558	0.814766
H	4.482848	0.602887	0.679044
H	2.146987	0.943970	0.710016
H	0.514477	-1.388018	-0.171036
H	-1.709956	-0.872369	-0.401645
H	-2.557604	1.895427	0.232129
H	-5.878620	0.474606	-0.250130
H	-2.684521	0.872018	-2.652331
H	-1.605520	2.144895	-2.032645
H	-3.355046	2.405377	-2.040432
H	3.705066	1.500602	-1.516349
H	4.257153	0.019894	-2.335331
H	5.436564	1.126341	-1.585742
H	-0.603949	-1.538210	2.057028
H	0.505263	-0.301991	2.701419
H	1.142041	-1.880330	2.182536
O	4.138239	-1.804514	2.343135
H	3.604852	-2.629766	2.277524
H	4.544396	-1.805226	3.221807
O	2.730606	-3.964477	1.454033
H	2.803440	-3.460640	0.621564
H	1.768597	-4.155062	1.514730
O	5.084044	-3.051562	-2.214715
H	5.637670	-2.935769	-3.000423
H	4.172872	-3.196059	-2.541134
O	2.282823	-3.172967	-2.758609
H	2.048647	-2.663866	-3.549957
H	2.299275	-2.534979	-2.012831
O	2.936681	2.664222	1.007156
H	2.480856	3.152583	0.275747
H	2.401179	2.895414	1.795171
O	7.806340	0.094261	-0.565678
H	8.537135	-0.108037	0.036175
H	7.720701	1.079129	-0.576684
O	7.404626	2.773917	-0.572957
H	7.159422	3.187519	-1.412783
H	6.694392	3.001025	0.074530
O	5.567015	3.314627	1.357474
H	4.613371	3.099114	1.226110
H	5.826395	2.887686	2.186661
O	-1.640332	-2.722837	-0.414012
H	-2.556412	-3.066666	-0.330999
H	-1.208057	-3.253552	-1.114823
O	0.050477	-4.485577	1.025944
H	-0.563363	-3.733205	0.922366
H	0.168123	-4.776125	0.099589
O	0.033845	-4.507305	-1.805435
H	0.836241	-4.136719	-2.237695
H	-0.275592	-5.230460	-2.370576
O	0.740725	3.213328	2.615893

H	0.515526	3.010928	3.535964
H	0.292190	2.545283	2.054836
O	1.261901	3.775580	-0.841445
H	0.697408	2.988723	-0.725681
H	0.783347	4.457650	-0.312655
O	-4.081291	-2.275483	2.454234
H	-3.477154	-2.448339	3.191807
H	-3.680759	-1.574791	1.903858
O	-4.201873	-3.741087	0.124326
H	-4.251141	-3.411985	1.049013
H	-4.262610	-4.707313	0.169746
O	-6.650762	-1.190506	2.620008
H	-7.302650	-1.839745	2.921799
H	-5.777977	-1.633997	2.677276
O	-7.012665	-0.383223	0.027456
H	-6.932087	-0.659279	0.973548
H	-6.799142	-1.194938	-0.498262
O	-6.195575	-2.551696	-1.412719
H	-5.460719	-2.932699	-0.881602
H	-5.785495	-2.224505	-2.226902
O	-0.230670	5.262315	0.932627
H	0.069785	4.727020	1.693173
H	-1.141222	4.944888	0.738734
O	-2.756395	4.246226	0.344623
H	-3.430670	4.467311	1.003311
H	-3.115129	4.563182	-0.518141
O	-5.871640	3.232679	-2.110713
H	-5.574338	2.485898	-1.556707
H	-6.093029	2.849206	-2.972181
O	-3.786406	5.008389	-2.077361
H	-4.074322	5.931946	-2.117363
H	-4.598927	4.460453	-2.175054

Table S20. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-β_B** conformation.

N	5.364325	-1.166673	-0.043761
C	4.322214	-0.111324	-0.109001
C	2.975379	-0.806946	0.033996
N	1.961647	-0.072859	0.460806
C	0.621537	-0.624770	0.597941
C	-0.376848	0.486948	0.301164
N	-1.559260	0.109227	-0.165634
C	-2.624449	1.067359	-0.409452
C	-3.944389	0.394463	-0.075876
O	-5.008644	1.099110	-0.400582
O	2.866804	-2.004306	-0.309508
O	-0.081693	1.674101	0.538956
C	-2.578763	1.610655	-1.839585
O	-4.012980	-0.703694	0.451530
C	4.409093	0.632919	-1.438807
C	0.390646	-1.172287	2.011545
H	6.313526	-0.746675	-0.165262
H	5.221452	-1.875561	-0.794479
H	5.297584	-1.632919	0.875663
H	4.483931	0.571566	0.728726
H	2.136115	0.904214	0.730601
H	0.510376	-1.432218	-0.132249
H	-1.732211	-0.888591	-0.346502
H	-2.499242	1.911926	0.280071

H	-5.876157	0.545129	-0.153821
H	-2.737378	0.810046	-2.568786
H	-1.598024	2.060774	-2.016552
H	-3.336800	2.382722	-1.984857
H	3.661474	1.429512	-1.468190
H	4.232798	-0.049354	-2.277185
H	5.398399	1.085280	-1.550798
H	-0.604939	-1.619595	2.091187
H	0.479115	-0.368545	2.749658
H	1.139775	-1.937575	2.232692
O	4.181481	-1.743052	2.434498
H	3.664078	-2.572222	2.482051
H	4.513878	-1.571651	3.327524
O	2.588609	-3.888086	1.647848
H	2.753357	-3.404553	0.810172
H	3.071546	-4.726280	1.580364
O	5.125910	-3.012557	-2.208675
H	5.718877	-2.846642	-2.955865
H	4.227198	-3.100180	-2.586257
O	2.345941	-2.998460	-2.778279
H	2.137355	-2.348383	-3.467108
H	2.371834	-2.501218	-1.932001
O	2.942242	2.626382	0.980610
H	2.473162	3.105228	0.250883
H	2.428684	2.878137	1.776737
O	7.800108	0.073302	-0.597350
H	8.549556	-0.105554	-0.011180
H	7.717632	1.057120	-0.649950
O	7.400815	2.750815	-0.698328
H	7.141754	3.142166	-1.544679
H	6.695155	2.983485	-0.048077
O	5.580348	3.285949	1.248964
H	4.623742	3.068803	1.144724
H	5.859263	2.876476	2.080630
O	-1.749434	-2.737818	-0.570103
H	-1.303110	-3.102119	-1.361391
H	-1.272900	-3.199696	0.156647
O	-0.056682	-4.415526	0.767714
H	0.785434	-4.236494	1.230766
H	0.190913	-4.628931	-0.154079
O	0.026708	-4.339340	-2.050144
H	0.846202	-3.962302	-2.441213
H	-0.234459	-5.081447	-2.615045
O	0.781777	3.227106	2.619909
H	0.566572	3.052156	3.547961
H	0.324993	2.544655	2.084089
O	1.249338	3.721812	-0.857894
H	0.677229	2.943749	-0.723146
H	0.788690	4.418234	-0.331405
O	-4.242256	-2.573522	2.373303
H	-3.672136	-2.856784	3.103318
H	-3.788794	-1.840201	1.914864
O	-4.351896	-3.730603	-0.215268
H	-4.455806	-3.520263	0.732759
H	-3.478162	-3.342829	-0.441442
O	-6.675488	-1.248116	2.648402
H	-7.387184	-1.823177	2.964622
H	-5.858946	-1.792916	2.653749
O	-7.031867	-0.286408	0.108244
H	-6.944509	-0.614523	1.036853
H	-6.863610	-1.078524	-0.464516
O	-6.388210	-2.414387	-1.450981
H	-5.608959	-2.862848	-1.033331
H	-6.096291	-2.128899	-2.328688

O	-0.188163	5.257367	0.914429
H	0.111213	4.727095	1.679073
H	-1.105043	4.951930	0.731505
O	-2.721553	4.261837	0.338401
H	-3.400668	4.485194	0.991280
H	-3.077904	4.566017	-0.530001
O	-5.833889	3.219726	-2.113914
H	-5.543129	2.488195	-1.536201
H	-6.046378	2.813187	-2.967003
O	-3.740494	4.985500	-2.100005
H	-4.023116	5.909626	-2.159209
H	-4.555145	4.440037	-2.194335

Table S21. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-β_C** conformation.

N	-5.105766	-1.488006	0.585286
C	-4.182753	-0.374757	0.249862
C	-2.808437	-0.986114	0.010268
N	-1.988829	-0.303666	-0.775454
C	-0.614914	-0.706547	-1.038129
C	0.303057	0.433521	-0.608357
N	1.453538	0.097246	-0.042501
C	2.427269	1.101122	0.354015
C	3.813463	0.549446	0.072481
O	4.797745	1.310372	0.508057
O	-2.505798	-2.052645	0.585931
O	-0.033944	1.613411	-0.827525
C	2.241672	1.519460	1.814527
O	4.002332	-0.504780	-0.510484
C	-4.139760	0.630059	1.398802
C	-0.412010	-0.997760	-2.526627
H	-6.067504	-1.113947	0.753157
H	-4.790762	-1.981378	1.448136
H	-5.115290	-2.155527	-0.202669
H	-4.547659	0.095735	-0.666988
H	-2.333392	0.569327	-1.197402
H	-0.405138	-1.598802	-0.441980
H	1.664315	-0.898003	0.124477
H	2.285634	1.989126	-0.274996
H	5.715680	0.841797	0.276974
H	2.387024	0.668103	2.486884
H	1.227974	1.907879	1.946375
H	2.945404	2.310378	2.082033
H	-3.454469	1.449394	1.165172
H	-3.804826	0.144430	2.321505
H	-5.136240	1.050281	1.562506
H	0.631116	-1.265254	-2.720470
H	-0.664770	-0.116859	-3.125593
H	-1.058082	-1.826227	-2.830703
O	-4.277440	-2.524972	-1.885348
H	-3.558576	-3.189198	-1.890447
H	-4.778686	-2.653621	-2.703385
O	-2.032676	-4.060908	-1.211278
H	-2.092536	-3.513278	-0.398600
H	-2.283754	-4.960365	-0.948212
O	-4.369705	-2.690220	3.058136
H	-4.848808	-2.342826	3.824530
H	-3.417333	-2.614378	3.273687
O	-1.570963	-2.333875	3.134517

H	-1.342316	-1.507071	3.587194
H	-1.727391	-2.095217	2.195884
O	-3.222762	2.209936	-1.546167
H	-2.886880	2.693380	-0.748924
H	-2.664024	2.564453	-2.269338
O	-7.530530	-0.264705	1.186127
H	-8.349250	-0.674479	0.871098
H	-7.550484	0.673911	0.874419
O	-7.423940	2.295042	0.319014
H	-7.108829	2.946410	0.961894
H	-6.844546	2.380557	-0.476176
O	-5.913376	2.451598	-1.940367
H	-4.936503	2.380867	-1.825487
H	-6.159872	1.771352	-2.583493
O	1.841429	-2.716235	0.370591
H	1.451819	-3.075814	1.201449
H	1.440089	-3.226833	-0.370209
O	0.697529	-4.112825	-1.703257
H	-0.283002	-4.111138	-1.651785
H	0.929155	-3.858728	-2.607987
O	0.839674	-3.680357	2.752284
H	-0.037629	-3.300256	2.973365
H	0.735046	-4.642051	2.788017
O	-0.980773	3.221233	-2.821335
H	-0.624223	3.192538	-3.721380
H	-0.494768	2.548979	-2.296921
O	-1.848792	3.259671	0.553466
H	-1.159453	2.590603	0.378178
H	-1.437857	4.080153	0.190285
O	4.260978	-2.259801	-2.506534
H	3.738979	-2.529988	-3.276351
H	3.741859	-1.593824	-2.016370
O	4.489579	-3.510625	0.035881
H	4.599989	-3.248758	-0.897879
H	3.587001	-3.184740	0.255044
O	6.628023	-0.772195	-2.574986
H	7.365035	-1.289493	-2.930438
H	5.835395	-1.347822	-2.635009
O	6.937713	0.097007	0.006282
H	6.868179	-0.200969	-0.933896
H	6.797954	-0.723080	0.546248
O	6.330029	-2.110155	1.468400
H	5.631513	-2.594841	0.957646
H	5.913460	-1.843220	2.300634
O	-0.421597	5.160512	-0.830248
H	-0.572283	4.678914	-1.667391
H	0.499223	4.937755	-0.566276
O	2.149104	4.384921	-0.082716
H	2.850168	4.724180	-0.657761
H	2.424635	4.619400	0.834921
O	5.310823	3.448833	2.321061
H	5.126015	2.723056	1.694477
H	5.586485	3.020150	3.144761
O	3.001879	4.921563	2.469005
H	3.163804	5.859880	2.644611
H	3.879742	4.477000	2.508525

Table S22. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 24H_2O$ peptide-water complex in the **$24H_2O$_β-β_A** conformation.

N	5.043648	-1.312494	-0.324804
C	4.044495	-0.220688	-0.456507
C	2.672262	-0.863692	-0.299746
N	1.698525	-0.090520	0.151855
C	0.358983	-0.603838	0.388161
C	-0.638730	0.505107	0.090228
N	-1.824133	0.122631	-0.365044
C	-2.906476	1.071628	-0.563333
C	-4.210826	0.354148	-0.266038
O	-5.289040	1.069272	-0.501268
O	2.520046	-2.058038	-0.626662
O	-0.346144	1.693400	0.325500
C	-2.878512	1.687578	-1.964499
O	-4.251690	-0.790710	0.160771
C	4.174263	0.450356	-1.819685
C	0.191861	-1.054562	1.844310
H	6.010687	-0.933143	-0.443125
H	4.883172	-2.054195	-1.040824
H	4.946876	-1.736941	0.611447
H	4.223359	0.499697	0.347698
H	1.925814	0.879370	0.406219
H	0.194221	-1.450785	-0.283038
H	-1.999246	-0.881053	-0.507262
H	-2.797912	1.883024	0.167904
H	-6.144660	0.501016	-0.243539
H	-3.030000	0.922963	-2.732462
H	-1.906039	2.160950	-2.124335
H	-3.648851	2.455104	-2.065124
H	3.464461	1.278357	-1.888339
H	3.969588	-0.266294	-2.622050
H	5.184368	0.850048	-1.945491
H	-0.816727	-1.447078	2.007277
H	0.359223	-0.212340	2.523356
H	0.920254	-1.838983	2.069777
O	3.858075	-1.848485	2.253493
H	3.283619	-2.649570	2.137166
H	4.453055	-2.048821	2.993063
O	2.411063	-3.946436	1.405265
H	2.466367	-3.522893	0.527441
H	1.447480	-4.112265	1.512901
O	4.732555	-3.335170	-2.315320
H	5.271954	-3.261276	-3.115951
H	3.814164	-3.485858	-2.618032
O	1.923201	-3.461501	-2.834555
H	1.685949	-3.029991	-3.669814
H	1.945933	-2.755875	-2.153347
O	2.790440	2.593843	0.377827
H	2.165660	3.130579	-0.169438
H	2.512211	2.758539	1.300317
O	7.552272	-0.196058	-0.809709
H	8.191979	-0.193938	-0.083005
H	7.517285	0.732091	-1.149853
O	7.288973	2.369708	-1.649397
H	7.019309	2.547486	-2.561634
H	6.568863	2.711120	-1.059170
O	5.507965	3.094540	0.219073
H	4.533680	2.997415	0.140562
H	5.737003	2.539511	0.996877
O	-1.946146	-2.739021	-0.453675
H	-2.865438	-3.065571	-0.341586
H	-1.526402	-3.322832	-1.118980
O	-0.279544	-4.434945	1.095355
H	-0.879670	-3.678709	0.948688
H	-0.187191	-4.794188	0.190196

O	-0.330493	-4.666253	-1.724138
H	0.470459	-4.355536	-2.203482
H	-0.678330	-5.418000	-2.226164
O	0.841686	3.154665	2.328337
H	0.644418	2.939180	3.251668
H	0.369614	2.492135	1.779380
O	0.806548	3.831694	-1.060940
H	0.255774	3.044072	-0.894948
H	0.410864	4.500958	-0.454584
O	-4.288776	-2.202196	2.448945
H	-3.662024	-2.363399	3.170249
H	-3.895871	-1.525849	1.863681
O	-4.502865	-3.726589	0.166822
H	-4.522349	-3.381272	1.086667
H	-4.570475	-4.691376	0.229814
O	-6.819567	-1.042602	2.683924
H	-7.473572	-1.636664	3.079818
H	-5.960551	-1.513905	2.724156
O	-7.276163	-0.341038	0.078421
H	-7.172112	-0.592525	1.029047
H	-7.081245	-1.166071	-0.434018
O	-6.514601	-2.538179	-1.346453
H	-5.772419	-2.918107	-0.824876
H	-6.115971	-2.217771	-2.169031
O	-0.446499	5.246410	0.940989
H	-0.041997	4.654853	1.606992
H	-1.364994	4.917757	0.817995
O	-2.988548	4.213920	0.452526
H	-3.654348	4.406117	1.128647
H	-3.361725	4.559695	-0.392869
O	-6.147719	3.310866	-2.039407
H	-5.852386	2.535638	-1.524846
H	-6.371534	2.973294	-2.919195
O	-4.049738	5.069377	-1.925824
H	-4.333540	5.995094	-1.920858
H	-4.865657	4.530306	-2.042833
O	5.787524	1.462070	2.429903
H	4.852428	1.210110	2.609878
H	6.281826	0.633370	2.349302
O	3.132122	0.780433	2.694614
H	3.167689	-0.199756	2.635716
H	2.733953	0.985598	3.553485

Table S23. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}27H_2O$ peptide-water complex in the **$27H_2O$_β-β_A** conformation.

N	-4.998333	-1.332625	-0.063772
C	-3.974030	-0.266863	0.087431
C	-2.617726	-0.936220	-0.079881
N	-1.630521	-0.180757	-0.524715
C	-0.289881	-0.709193	-0.693940
C	0.699947	0.418367	-0.463588
N	1.883614	0.074769	0.028637
C	2.956397	1.044711	0.175708
C	4.269486	0.310812	-0.027442
O	5.340777	1.051081	0.148921
O	-2.489443	-2.137989	0.231199
O	0.411434	1.587737	-0.786381
C	2.887636	1.773933	1.519615
O	4.318857	-0.871536	-0.334890

C	-4.082152	0.375195	1.466343
C	-0.090502	-1.271486	-2.106992
H	-5.958325	-0.929368	0.028650
H	-4.877104	-2.066948	0.667124
H	-4.888658	-1.768941	-0.992710
H	-4.137085	0.474403	-0.700746
H	-1.829178	0.798162	-0.760435
H	-0.139057	-1.497514	0.048822
H	2.069757	-0.918706	0.225181
H	2.865376	1.794674	-0.619873
H	6.202022	0.459813	-0.038973
H	3.007853	1.074450	2.353598
H	1.917583	2.273090	1.608749
H	3.662182	2.540851	1.584593
H	-3.350243	1.182449	1.551211
H	-3.888632	-0.366813	2.248751
H	-5.082433	0.794345	1.608742
H	0.918571	-1.681330	-2.216288
H	-0.236133	-0.482029	-2.851283
H	-0.818546	-2.066893	-2.287687
O	-3.753277	-1.914638	-2.618048
H	-3.198831	-2.726470	-2.494748
H	-4.337542	-2.102457	-3.369447
O	-2.410368	-4.094561	-1.729161
H	-2.548058	-3.665944	-0.863493
H	-1.441043	-4.256826	-1.729963
O	-4.804771	-3.241303	2.045179
H	-5.381472	-3.079077	2.805708
H	-3.903239	-3.382785	2.406542
O	-2.055570	-3.372456	2.597387
H	-1.855310	-2.705209	3.298710
H	-2.084352	-2.852844	1.765183
O	-2.708886	2.536228	-0.687889
H	-2.092386	3.045679	-0.114938
H	-2.411283	2.738990	-1.597643
O	-7.498565	-0.183042	0.380921
H	-8.119575	-0.139915	-0.360620
H	-7.457924	0.729424	0.760361
O	-7.216676	2.347282	1.314902
H	-6.935129	2.493757	2.229035
H	-6.491470	2.682038	0.727313
O	-5.425051	3.053760	-0.550246
H	-4.452646	2.954624	-0.459140
H	-5.642927	2.501947	-1.333402
O	1.981285	-2.770175	0.364130
H	2.896055	-3.125513	0.334775
H	1.513430	-3.274780	1.062110
O	0.244510	-4.510389	-1.070842
H	0.891115	-3.784141	-0.986015
H	0.106030	-4.765627	-0.136252
O	0.278608	-4.523830	1.757928
H	-0.550586	-4.201459	2.188315
H	0.602256	-5.264124	2.291982
O	-0.816592	3.192749	-2.653214
H	-0.719020	3.083055	-3.610468
H	-0.321097	2.459550	-2.233526
O	-0.591552	3.638024	0.728131
H	-0.111090	2.874394	0.343143
H	-0.239084	4.400769	0.205888
O	4.424935	-2.543663	-2.446036
H	3.804698	-2.771316	-3.154796
H	4.031458	-1.807143	-1.938801
O	4.564608	-3.813559	-0.013150
H	4.617473	-3.580573	-0.966226

H	4.645099	-4.777785	0.043309
O	6.962689	-1.420881	-2.767583
H	7.626247	-2.073509	-3.034447
H	6.101699	-1.890548	-2.769737
O	7.333466	-0.406311	-0.251609
H	7.253911	-0.757386	-1.172687
H	7.130660	-1.175839	0.338757
O	6.564161	-2.455931	1.372945
H	5.823367	-2.890718	0.894302
H	6.167612	-2.061494	2.163651
O	0.516113	5.209312	-1.181321
H	0.085864	4.651547	-1.860575
H	1.442038	4.882050	-1.119705
O	3.061104	4.161215	-0.822045
H	3.698236	4.345502	-1.527463
H	3.459273	4.535042	-0.000005
O	6.211116	3.287922	1.694294
H	5.907091	2.513898	1.183273
H	6.400355	2.957721	2.584960
O	4.163363	5.096702	1.505560
H	4.465807	6.015335	1.459400
H	4.967939	4.546456	1.646574
O	-5.655816	1.440478	-2.783553
H	-4.721854	1.181584	-2.960074
H	-6.161090	0.615879	-2.735491
O	-3.007988	0.722405	-3.057334
H	-3.046818	-0.256346	-2.983696
H	-2.623097	0.910365	-3.926291
O	-1.436321	-1.327007	4.283844
H	-1.066950	-0.705543	3.612092
H	-2.206074	-0.884036	4.669195
O	-0.324354	0.248674	2.326481
H	-0.312250	1.166900	2.692776
H	0.605982	-0.024161	2.292446
O	-0.313280	2.813536	3.289268
H	-0.472406	3.323480	2.462299
H	-1.063569	2.995968	3.873243

Table S24. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 31H_2O$ peptide-water complex in the **$31H_2O$_β-β_A** conformation.

N	5.557723	0.922264	-0.097785
C	4.396831	0.012583	0.091133
C	3.149172	0.827450	-0.220557
N	2.148301	0.171361	-0.785034
C	0.874338	0.792418	-1.102125
C	-0.213795	-0.147001	-0.602803
N	-1.194763	0.391183	0.120443
C	-2.310248	-0.415343	0.580340
C	-3.581155	0.381768	0.388848
O	-4.677184	-0.319039	0.642234
O	3.128384	2.036706	0.075076
O	-0.157362	-1.357112	-0.877626
C	-2.119041	-0.860655	2.033607
O	-3.605917	1.551439	0.051701
C	4.351787	-0.498052	1.526090
C	0.719859	0.998744	-2.610696
H	6.449830	0.413468	0.094015
H	5.496731	1.741316	0.548171
H	5.553595	1.266692	-1.070343

H	4.503796	-0.817406	-0.613784
H	2.271637	-0.831189	-0.968351
H	0.831800	1.750247	-0.574003
H	-1.223338	1.411434	0.259599
H	-2.372168	-1.304545	-0.055474
H	-5.492448	0.251580	0.400422
H	-2.087050	0.001279	2.708714
H	-1.177471	-1.412244	2.119874
H	-2.923250	-1.529726	2.348130
H	3.515843	-1.194556	1.634610
H	4.213002	0.332734	2.226849
H	5.280023	-1.023716	1.767939
H	-0.241083	1.471100	-2.837419
H	0.771039	0.035926	-3.128658
H	1.525100	1.640867	-2.977429
O	4.498096	1.346661	-2.774999
H	3.957440	2.176519	-2.751477
H	5.131188	1.461400	-3.501198
O	3.125909	3.606841	-2.186963
H	3.165807	3.291038	-1.262113
H	2.179965	3.844630	-2.288932
O	5.511242	3.067830	1.751870
H	6.051798	2.936897	2.544258
H	4.598388	3.250782	2.068864
O	2.821998	3.364876	2.390676
H	2.522286	3.052725	3.275225
H	2.663176	2.614484	1.780265
O	2.785473	-2.645763	-0.574526
H	2.021730	-2.944947	-0.027839
H	2.543662	-2.913933	-1.482683
O	7.853920	-0.505984	0.609371
H	8.487714	-0.716741	-0.091518
H	7.663058	-1.358156	1.073132
O	7.163892	-2.859732	1.774988
H	6.831517	-2.868571	2.683764
H	6.421182	-3.154461	1.188140
O	5.367599	-3.514313	-0.103979
H	4.414685	-3.278543	-0.104423
H	5.708376	-3.094841	-0.924675
O	-1.098294	3.219990	0.363753
H	-1.989862	3.615456	0.467433
H	-0.474558	3.793676	0.858728
O	0.541963	4.428269	-1.626358
H	-0.187480	3.852953	-1.328899
H	0.849110	4.816461	-0.782505
O	0.964368	4.922905	1.136067
H	1.672081	4.468605	1.656134
H	0.795453	5.769299	1.575739
O	0.873307	-3.342299	-2.499208
H	0.763825	-3.273516	-3.459002
H	0.453931	-2.544782	-2.113013
O	0.529206	-3.425823	0.825376
H	-0.123174	-2.849976	0.373994
H	0.327862	-4.321364	0.472251
O	-3.358898	3.022296	-2.186634
H	-2.727060	3.225477	-2.892877
H	-2.981736	2.292512	-1.656550
O	-3.651941	4.414212	0.174706
H	-3.662416	4.120242	-0.762439
H	-3.655377	5.383323	0.158562
O	-5.579247	1.506683	-2.613884
H	-6.140889	1.613020	-3.396745
H	-4.929838	2.245480	-2.618572
O	-6.641485	1.187215	-0.098198

H	-6.361023	1.381075	-1.023457
H	-6.466924	2.016010	0.422045
O	-5.939970	3.353787	1.360402
H	-5.095903	3.674564	0.969201
H	-5.715151	3.045532	2.250553
O	-0.357253	-5.367056	-0.905708
H	0.056003	-4.815784	-1.600546
H	-1.209449	-4.916889	-0.707496
O	-2.308361	-3.692028	0.036496
H	-3.130122	-3.441126	-0.449833
H	-2.588273	-3.879517	0.958103
O	-5.413596	-2.392531	2.410521
H	-5.109674	-1.590680	1.940279
H	-5.833814	-2.087841	3.229190
O	-3.236270	-4.118385	2.642420
H	-3.501893	-5.036806	2.795705
H	-4.064807	-3.593636	2.649465
O	5.961581	-2.230181	-2.478230
H	5.086324	-1.881554	-2.766426
H	6.570333	-1.477095	-2.492086
O	3.474336	-1.211258	-3.068248
H	3.655280	-0.244543	-3.066845
H	3.120285	-1.419238	-3.945648
O	1.743222	2.089470	4.566360
H	1.515494	1.415604	3.887557
H	2.340769	1.655095	5.192003
O	1.312256	0.677619	2.240054
H	1.304531	-0.291510	2.429364
H	0.408478	0.887400	1.955413
O	1.247188	-1.952936	3.002957
H	1.042140	-2.624112	2.315324
H	2.030708	-2.271573	3.474137
O	-4.483819	-2.855476	-1.394806
H	-5.331779	-2.816469	-0.896124
H	-4.249419	-1.935925	-1.637427
O	-6.837363	-2.988402	0.058570
H	-7.465168	-2.237915	-0.026911
H	-6.505039	-2.939112	0.975054
O	-3.540904	-0.358587	-2.253768
H	-3.034292	-0.550661	-3.057105
H	-4.300489	0.192790	-2.539274
O	-8.564715	-0.801126	0.000667
H	-9.199683	-0.746028	-0.727786
H	-8.011072	0.006255	-0.055723

Table S25. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 37H_2O$ peptide-water complex in the **$37H_2O$_β-β_A** conformation.

N	-4.531983	-0.233048	-0.060667
C	-3.287196	0.539594	0.194690
C	-2.127198	-0.377162	-0.162344
N	-1.090856	0.193704	-0.751439
C	0.114286	-0.533807	-1.109525
C	1.297404	0.284291	-0.613791
N	2.216875	-0.354864	0.104106
C	3.418863	0.316085	0.561740
C	4.582848	-0.632806	0.379838
O	5.757182	-0.065833	0.617838
O	-2.209998	-1.592937	0.101170
O	1.372184	1.493577	-0.895342

C	3.284921	0.787104	2.013448
O	4.463351	-1.802985	0.066361
C	-3.215705	0.967924	1.654127
C	0.213325	-0.716639	-2.625532
H	-5.344617	0.407076	0.036863
H	-4.628311	-1.037692	0.599084
H	-4.499077	-0.593130	-1.031470
H	-3.295339	1.414605	-0.462096
H	-1.129865	1.206593	-0.910154
H	0.077359	-1.503868	-0.604516
H	2.109669	-1.361879	0.285588
H	3.587846	1.188715	-0.077179
H	6.495229	-0.738052	0.391279
H	3.148183	-0.060476	2.693647
H	2.419562	1.451077	2.098738
H	4.165875	1.353808	2.323727
H	-2.336524	1.600461	1.801523
H	-3.139069	0.095863	2.312908
H	-4.109956	1.540716	1.916211
H	1.129969	-1.253619	-2.889154
H	0.220710	0.258697	-3.122827
H	-0.648074	-1.288789	-2.979532
O	-3.625252	-0.731902	-2.714196
H	-3.121281	-1.575028	-2.765292
H	-4.425286	-0.831410	-3.289705
O	-2.344751	-3.117342	-2.202242
H	-2.370720	-2.776402	-1.286336
H	-1.443420	-3.498076	-2.257146
O	-4.989816	-2.299369	1.792379
H	-5.429146	-1.805274	2.523357
H	-4.160902	-2.698800	2.136668
O	-2.391429	-3.081513	2.348499
H	-2.077377	-2.677140	3.191928
H	-2.207660	-2.409717	1.655297
O	-1.439210	3.061800	-0.404786
H	-0.633817	3.258507	0.128666
H	-1.191701	3.338251	-1.309279
O	-6.314846	1.930407	-0.066593
H	-5.917663	2.385304	-0.843500
H	-6.187761	2.536447	0.704969
O	-5.710909	3.743508	1.869081
H	-5.460532	3.492996	2.769573
H	-4.889045	4.044647	1.407434
O	-3.767154	4.432754	0.141688
H	-2.898809	3.974233	0.090943
H	-4.209265	4.186183	-0.695654
O	1.745086	-3.129833	0.506621
H	2.582364	-3.632677	0.602274
H	1.058454	-3.628335	0.999834
O	0.083272	-4.277892	-1.503818
H	0.840636	-3.753213	-1.183536
H	-0.294849	-4.615241	-0.667266
O	-0.448861	-4.675962	1.264844
H	-1.182465	-4.214602	1.739863
H	-0.325400	-5.529069	1.706630
O	0.400472	3.541130	-2.453250
H	0.459305	3.491010	-3.418674
H	0.781972	2.707759	-2.103811
O	0.952431	3.553947	0.895652
H	1.478268	2.894170	0.395759
H	1.239994	4.413198	0.511593
O	3.939355	-3.256643	-2.120731
H	3.309719	-3.396134	-2.844277
H	3.652506	-2.463869	-1.626596

O	4.121230	-4.623082	0.274963
H	4.146583	-4.338643	-0.664620
H	3.997222	-5.584365	0.266991
O	6.358764	-2.078922	-2.564995
H	6.887376	-2.293249	-3.348844
H	5.608672	-2.714769	-2.536602
O	7.507561	-1.829665	-0.084200
H	7.179738	-2.012458	-0.996480
H	7.231184	-2.609400	0.466054
O	6.531540	-3.843781	1.436514
H	5.652818	-4.058975	1.049119
H	6.348448	-3.492636	2.320360
O	1.962508	5.368356	-0.894973
H	1.456380	4.883033	-1.576993
H	2.773751	4.830155	-0.748092
O	3.820656	3.557317	-0.036667
H	4.600544	3.211520	-0.533025
H	4.131348	3.723980	0.878841
O	6.761663	1.947637	2.322854
H	6.350526	1.179742	1.878490
H	7.164040	1.609807	3.137517
O	4.795656	3.905479	2.564981
H	5.155748	4.789744	2.726382
H	5.563202	3.294762	2.573520
O	-4.883471	3.158288	-2.100714
H	-4.094738	2.679556	-2.463170
H	-5.336093	3.563201	-2.855603
O	-2.644085	1.802769	-2.826737
H	-2.877559	0.846993	-2.914698
H	-2.225564	2.060996	-3.661274
O	-1.335554	-1.650436	4.421107
H	-0.920606	-1.057660	3.751552
H	-1.966982	-1.106551	4.913749
O	-0.297268	-0.334893	2.256139
H	-0.075569	0.594354	2.504488
H	0.549966	-0.759174	2.049100
O	0.207910	2.210930	3.147297
H	0.437866	2.853697	2.441021
H	-0.548702	2.583908	3.622747
O	5.853116	2.445952	-1.496246
H	6.690274	2.298468	-0.999812
H	5.495964	1.562866	-1.724859
O	8.217196	2.286540	-0.062264
H	8.745281	1.462123	-0.141427
H	7.893217	2.297972	0.858500
O	4.598118	0.064189	-2.283956
H	4.102458	0.277963	-3.088568
H	5.274333	-0.595042	-2.549793
O	9.667600	-0.096191	-0.127577
H	10.228210	-0.243676	-0.902710
H	9.013512	-0.826760	-0.121627
O	-8.666179	0.667297	-0.404870
H	-9.415978	1.226777	-0.656130
H	-7.881088	1.260335	-0.293538
O	-7.241433	-2.951320	0.272675
H	-7.851695	-2.440524	0.843181
H	-6.377896	-2.899378	0.730318
O	-5.992584	-0.846518	-4.007420
H	-6.121746	-1.474834	-4.732144
H	-6.633284	-1.103298	-3.296060
O	-7.700373	-1.449568	-2.001982
H	-7.423888	-2.042061	-1.266114
H	-8.090334	-0.667425	-1.562126
O	-6.532925	-0.687568	3.353767

H	-7.387683	-0.759042	2.874251
H	-6.728236	-0.886553	4.280933
O	-8.743363	-1.058575	1.713387
H	-9.624537	-1.212427	2.085388
H	-8.842809	-0.372077	1.011481

Table S26. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **β-PPII** conformation.

N	5.176226	0.122226	-0.437200
C	3.909605	-0.676915	-0.403168
C	2.828814	0.301244	0.072127
N	1.570645	-0.028614	-0.225766
C	0.442727	0.756360	0.245271
C	-0.789631	-0.137928	0.152561
N	-1.845227	0.233883	0.901918
C	-3.121770	-0.427505	0.752326
C	-3.677945	-0.183133	-0.647464
O	-4.635561	-1.070152	-0.941275
O	3.153581	1.288175	0.733542
O	-0.816372	-1.115720	-0.592637
C	-4.110652	0.078908	1.802711
O	-3.336518	0.706556	-1.389690
C	4.056342	-1.839522	0.572916
C	0.239813	2.018673	-0.602095
H	6.011531	-0.464555	-0.346079
H	5.150495	0.798964	0.342426
H	5.270250	0.664559	-1.302661
H	3.716228	-1.030767	-1.417112
H	1.349350	-0.841197	-0.796337
H	0.629592	1.032853	1.288193
H	-1.797114	1.082161	1.454448
H	-2.978779	-1.506979	0.864546
H	-5.000800	-0.861754	-1.821277
H	-4.277757	1.154823	1.689168
H	-3.715004	-0.115438	2.802660
H	-5.066934	-0.436908	1.698219
H	3.118949	-2.398158	0.614354
H	4.289692	-1.475800	1.578048
H	4.844290	-2.520327	0.240975
H	-0.587877	2.617879	-0.212750
H	0.020377	1.744039	-1.638095
H	1.150142	2.622137	-0.576675

Table S27. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 2H_2O$ peptide-water complex in the **$2H_2O$_β-PPII** conformation.

N	4.547775	-0.893612	-0.282809
C	3.199674	-1.523111	-0.379933
C	2.241012	-0.550246	0.304966
N	0.983103	-0.557143	-0.130399
C	-0.047783	0.282606	0.455354
C	-1.386732	-0.382720	0.151342
N	-2.419809	-0.017054	0.933886
C	-3.766611	-0.431452	0.609487

C	-4.187132	0.160080	-0.732850
O	-5.241283	-0.494928	-1.232606
O	2.652148	0.153354	1.235713
O	-1.508192	-1.174027	-0.781823
C	-4.739539	0.010372	1.702770
O	-3.671375	1.110995	-1.270158
C	3.190184	-2.883613	0.308138
C	-0.019748	1.695017	-0.143832
H	5.278481	-1.501687	-0.662882
H	4.767736	-0.687905	0.698663
H	4.540792	0.022851	-0.799336
H	2.970926	-1.614476	-1.444267
H	0.691451	-1.153540	-0.901568
H	0.120439	0.327057	1.535977
H	-2.284552	0.685794	1.651382
H	-3.789869	-1.521230	0.508686
H	-5.512410	-0.068298	-2.066683
H	-4.743852	1.100609	1.799887
H	-4.442973	-0.430233	2.657832
H	-5.750515	-0.324171	1.462887
H	2.203936	-3.340941	0.201852
H	3.415802	-2.779519	1.373714
H	3.926870	-3.546784	-0.152131
H	-0.783888	2.327028	0.317298
H	-0.205444	1.646612	-1.221187
H	0.958834	2.150626	0.029153
O	4.074556	1.532709	-1.461480
H	3.750807	2.132757	-0.748762
H	3.413195	1.555893	-2.168677
O	3.185422	2.840179	0.773470
H	2.991654	2.002220	1.236921
H	3.874943	3.284133	1.288921

Table S28. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_β-PPII** conformation.

N	3.834839	0.505844	-1.425044
C	2.552490	-0.172177	-1.762705
C	1.675527	-0.105731	-0.516979
N	0.376442	0.088091	-0.713827
C	-0.590832	0.077681	0.371818
C	-1.942762	-0.256114	-0.254035
N	-2.872265	-0.767862	0.574162
C	-4.235812	-0.939235	0.124078
C	-4.857633	0.418536	-0.190070
O	-5.951339	0.277188	-0.947897
O	2.182445	-0.278730	0.603557
O	-2.164019	-0.028465	-1.441749
C	-5.063669	-1.661552	1.186973
O	-4.450297	1.483534	0.208929
C	2.809756	-1.625397	-2.150006
C	-0.662627	1.442337	1.067868
H	4.439181	0.565880	-2.249390
H	4.358611	-0.021272	-0.680923
H	3.636481	1.472526	-1.082368
H	2.096983	0.381052	-2.587269
H	-0.002575	0.237862	-1.646199
H	-0.294655	-0.696957	1.085824
H	-2.666025	-0.882755	1.559703
H	-4.232152	-1.519931	-0.803788

H	-6.346214	1.155892	-1.100429
H	-5.091627	-1.083366	2.115825
H	-4.624659	-2.641239	1.390503
H	-6.086127	-1.804145	0.831888
H	1.865088	-2.116540	-2.394560
H	3.285021	-2.161750	-1.322838
H	3.459713	-1.673691	-3.027366
H	-1.390218	1.422882	1.883839
H	-0.956510	2.213868	0.349724
H	0.317387	1.695795	1.480556
O	2.865541	2.868653	-0.331989
H	2.818413	2.696357	0.635912
H	1.991287	3.181833	-0.606394
O	3.005817	1.857277	2.192577
H	2.673088	0.978273	1.931064
H	2.542481	2.104130	3.005957
O	5.297009	-0.928649	0.454081
H	6.026774	-1.449626	0.088391
H	4.743254	-1.550782	0.980716
O	3.457347	-2.398925	1.859680
H	3.282672	-3.311870	1.588261
H	2.734549	-1.860090	1.487168

Table S29. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_β-PPII** conformation.

N	3.801366	-0.541870	0.287298
C	2.597000	-0.085427	-0.451730
C	1.395994	-0.834920	0.114064
N	0.264841	-0.141023	0.183114
C	-0.978579	-0.678051	0.703136
C	-2.063723	-0.604323	-0.364912
N	-3.039104	-1.527247	-0.285812
C	-4.268429	-1.348565	-1.028143
C	-4.953028	-0.061639	-0.575943
O	-5.878698	0.330715	-1.446537
O	1.518402	-2.026355	0.453018
O	-2.086064	0.298469	-1.205449
C	-5.201100	-2.542112	-0.823214
O	-4.710621	0.518757	0.460421
C	2.761032	-0.365612	-1.942653
C	-1.440079	0.144221	1.913201
H	4.623433	0.015344	-0.035805
H	3.998526	-1.550631	0.104392
H	3.647619	-0.392691	1.299888
H	2.491128	0.987946	-0.266068
H	0.278174	0.846742	-0.100195
H	-0.799885	-1.715903	0.993525
H	-3.023140	-2.208385	0.465155
H	-4.033471	-1.236890	-2.091195
H	-6.322022	1.131833	-1.108709
H	-5.466545	-2.651142	0.232932
H	-4.705049	-3.454981	-1.161394
H	-6.116210	-2.405924	-1.402453
H	1.887827	-0.001111	-2.489451
H	2.865538	-1.440883	-2.120290
H	3.651392	0.143629	-2.321552
H	-2.402164	-0.220524	2.283569
H	-1.553697	1.196358	1.629005

H	-0.696345	0.061732	2.708906
O	2.589841	0.042125	2.761643
H	1.927817	-0.632230	3.035329
H	2.184291	0.906797	2.919499
O	0.818214	-2.039324	3.122217
H	1.042803	-2.406503	2.245027
H	1.077319	-2.707704	3.773611
O	4.517429	-3.182311	-0.411297
H	5.170358	-3.183977	-1.126244
H	3.740827	-3.688804	-0.742110
O	2.091754	-4.305127	-1.021913
H	1.784169	-4.307702	-1.940033
H	1.656333	-3.543668	-0.590795
O	0.442092	2.647114	-0.588013
H	0.628908	2.705219	-1.538444
H	-0.506123	2.933321	-0.490859
O	5.719896	1.056290	-0.931158
H	6.564434	1.241128	-0.495285
H	5.308226	1.934582	-1.124704
O	4.435262	3.389631	-1.442746
H	3.946333	3.377204	-2.278252
H	3.775799	3.597953	-0.739167
O	2.606232	3.917567	0.530661
H	1.756270	3.504497	0.259824
H	2.842617	3.525082	1.383574
O	-2.203931	3.027220	-0.546462
H	-2.728815	3.125618	0.277943
H	-2.352325	2.105513	-0.835318
O	-3.923713	2.969846	1.629126
H	-4.345834	2.121851	1.398616
H	-3.571526	2.859913	2.524409

Table S30 M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_β-PPII_A** conformation.

N	-3.928408	0.458248	0.035609
C	-2.807435	-0.381079	-0.464000
C	-1.526575	0.408315	-0.231896
N	-0.424992	-0.289041	0.015953
C	0.856184	0.351374	0.246727
C	1.917646	-0.288909	-0.638266
N	2.974749	0.470778	-0.923765
C	4.156727	-0.115868	-1.515169
C	4.719322	-1.177949	-0.575664
O	5.557752	-1.994804	-1.207724
O	-1.560180	1.648270	-0.332024
O	1.837139	-1.475484	-1.003436
C	5.211376	0.960681	-1.774541
O	4.465953	-1.254084	0.607151
C	-2.981389	-0.670285	-1.952471
C	1.278542	0.203910	1.714126
H	-4.838514	-0.003149	-0.183648
H	-3.912938	1.409626	-0.388840
H	-3.817913	0.558969	1.060673
H	-2.805498	-1.302730	0.125397
H	-0.510689	-1.298411	0.200434
H	0.749953	1.410600	-0.004137
H	3.012606	1.427420	-0.553968
H	3.890743	-0.615092	-2.452360
H	5.929573	-2.633664	-0.570770

H	5.510966	1.442100	-0.838431
H	4.801871	1.717666	-2.447875
H	6.093555	0.518150	-2.240881
H	-2.169120	-1.315668	-2.298371
H	-2.969327	0.263363	-2.524587
H	-3.934335	-1.177519	-2.126351
H	2.250361	0.677209	1.882067
H	1.353145	-0.856113	1.982182
H	0.528259	0.677282	2.354372
O	-2.629184	0.357442	2.498274
H	-2.199467	1.234166	2.640455
H	-2.998863	0.085081	3.350500
O	-1.568667	2.825547	2.225042
H	-1.580171	2.655729	1.264339
H	-0.647629	3.117462	2.409266
O	-4.028830	3.167277	-0.907771
H	-4.734547	3.541761	-1.454105
H	-3.193386	3.564731	-1.224306
O	-1.358236	3.945549	-1.671803
H	-1.230239	4.028416	-2.629348
H	-1.207300	3.003662	-1.449931
O	-0.829956	-3.109202	0.434512
H	-0.781601	-3.282962	-0.531929
H	0.051169	-3.386396	0.784939
O	-6.145278	-1.032685	-0.782904
H	-6.978551	-0.981038	-0.292759
H	-5.845647	-1.973125	-0.729709
O	-5.061768	-3.512283	-0.606272
H	-4.588360	-3.814581	-1.394714
H	-4.401638	-3.494019	0.127596
O	-3.322615	-3.275217	1.476836
H	-2.378133	-3.352611	1.206101
H	-3.461700	-3.915873	2.189421
O	0.837814	4.948666	-0.287582
H	1.048617	5.858433	-0.545210
H	0.040006	4.689446	-0.803740
O	2.799486	3.064448	0.291383
H	3.656182	3.491557	0.446462
H	2.238806	3.741792	-0.147682
O	0.964718	3.880298	2.321977
H	1.683753	3.306746	2.000657
H	0.813056	4.472408	1.557732
O	1.807628	-3.596915	0.883807
H	2.319736	-3.353653	1.684865
H	2.030405	-2.912574	0.224126
O	-0.295532	-2.813965	-2.228503
H	0.502052	-2.314721	-1.957281
H	0.010886	-3.503159	-2.835974
O	3.545304	-2.695887	2.857401
H	4.028734	-2.099877	2.257275
H	3.220932	-2.143653	3.583469

Table S31. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_β-PPII_B** conformation.

N	-3.952926	0.436203	0.188396
C	-2.809021	-0.360844	-0.324247
C	-1.549906	0.458233	-0.084417
N	-0.406673	-0.206129	-0.015782
C	0.858323	0.473245	0.199177

C	1.939338	-0.195315	-0.638685
N	3.024955	0.538148	-0.872541
C	4.212323	-0.069974	-1.428832
C	4.714246	-1.160213	-0.487532
O	5.564970	-1.979825	-1.099782
O	-1.633600	1.703390	-0.031474
O	1.840258	-1.379162	-1.010631
C	5.299886	0.985661	-1.634412
O	4.404934	-1.258576	0.680696
C	-2.984196	-0.642906	-1.814463
C	1.258004	0.415340	1.680268
H	-4.845753	-0.073191	0.004471
H	-3.997005	1.370566	-0.272557
H	-3.823782	0.570383	1.204008
H	-2.773608	-1.288194	0.254682
H	-0.436425	-1.233419	0.052851
H	0.742048	1.515596	-0.112940
H	3.055339	1.510085	-0.529039
H	3.970948	-0.550795	-2.382681
H	5.891980	-2.642421	-0.462521
H	5.563320	1.458229	-0.683313
H	4.935394	1.753771	-2.320947
H	6.193308	0.528091	-2.063313
H	-2.159182	-1.265414	-2.172509
H	-2.996626	0.294257	-2.381637
H	-3.926165	-1.170835	-1.987377
H	2.210858	0.925598	1.845008
H	1.357351	-0.627129	2.002701
H	0.484342	0.899992	2.282715
O	-2.501190	0.304417	2.579479
H	-2.086553	1.143337	2.864325
H	-2.672529	-0.207617	3.383158
O	-1.313127	2.823290	2.448410
H	-1.463060	2.621044	1.498792
H	-1.953890	3.513674	2.680645
O	-4.235123	2.963579	-1.109827
H	-4.880433	3.010773	-1.829930
H	-3.406484	3.353055	-1.457334
O	-1.593214	3.716836	-1.823579
H	-1.388264	3.586582	-2.762128
H	-1.413145	2.862753	-1.378551
O	-0.781163	-3.053071	0.189976
H	-0.713577	-3.196470	-0.779956
H	0.088303	-3.349732	0.554261
O	-6.139966	-1.106794	-0.596509
H	-6.969002	-1.080607	-0.097126
H	-5.840131	-2.049068	-0.595469
O	-5.070318	-3.596136	-0.571922
H	-4.649202	-3.860616	-1.402451
H	-4.358376	-3.588454	0.112409
O	-3.182630	-3.399659	1.381149
H	-2.267350	-3.410655	1.013404
H	-3.222451	-4.105633	2.042600
O	0.726706	4.888117	-0.624188
H	0.608069	4.812341	0.342396
H	-0.108190	4.586697	-1.033462
O	2.840024	3.199682	0.039039
H	2.487654	3.332317	0.940310
H	2.210705	3.721979	-0.502164
O	1.120369	4.085181	2.075786
H	0.307041	3.618252	2.370410
H	1.369468	4.684643	2.794587
O	1.826651	-3.586813	0.733605
H	2.332345	-3.439620	1.561390

H	2.069006	-2.842028	0.150631
O	-0.199329	-2.652926	-2.448687
H	0.596410	-2.177281	-2.135302
H	0.107983	-3.295463	-3.104841
O	3.592272	-2.953092	2.790548
H	4.076467	-2.322245	2.226907
H	3.269164	-2.441544	3.546503

Table S32. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-PPII_A** conformation.

N	-4.565965	-0.985325	0.060767
C	-3.142884	-1.277141	-0.250866
C	-2.378004	0.027513	-0.077159
N	-1.082969	-0.070661	0.193538
C	-0.240227	1.097595	0.369488
C	1.037709	0.917030	-0.439161
N	1.719412	2.023877	-0.722858
C	3.040963	1.932211	-1.304328
C	3.965987	1.144783	-0.379435
O	4.990289	0.623275	-1.029394
O	-2.976084	1.107109	-0.239146
O	1.453130	-0.212143	-0.757146
C	3.616306	3.330444	-1.531862
O	3.798541	1.057986	0.823789
C	-3.003463	-1.798007	-1.679114
C	0.112363	1.297561	1.849146
H	-5.152124	-1.828722	-0.126237
H	-4.926031	-0.186084	-0.502066
H	-4.621280	-0.728430	1.061536
H	-2.799742	-2.018515	0.475865
H	-0.688362	-1.002061	0.381928
H	-0.792249	1.967641	0.001548
H	1.342057	2.932540	-0.432222
H	2.986509	1.391327	-2.255229
H	5.693767	0.229175	-0.396751
H	3.699655	3.871773	-0.584186
H	2.963394	3.891976	-2.204922
H	4.606569	3.258444	-1.985955
H	-1.957333	-2.037472	-1.891137
H	-3.346543	-1.046849	-2.398346
H	-3.599609	-2.706136	-1.805149
H	0.735731	2.186979	1.976857
H	0.657283	0.427938	2.232394
H	-0.810544	1.417993	2.424569
O	-3.543194	-0.297597	2.573840
H	-3.533004	0.681686	2.686309
H	-3.778241	-0.675311	3.433436
O	-3.607976	2.380428	2.177423
H	-3.492337	2.138118	1.239333
H	-2.917779	3.062418	2.334211
O	-5.728097	1.235853	-1.339891
H	-6.409686	1.170160	-2.023973
H	-5.124908	1.952372	-1.619777
O	-3.586593	3.093848	-1.921332
H	-3.347582	3.082853	-2.861031
H	-3.100555	2.352656	-1.502923
O	-0.257206	-2.822635	0.531667
H	-0.079838	-2.935371	-0.435164
H	0.641252	-2.768915	0.924513

O	-5.876838	-3.343046	-0.666154
H	-6.707354	-3.578630	-0.227971
H	-5.252833	-4.093078	-0.504611
O	-3.981487	-5.222967	-0.200150
H	-3.457882	-5.470291	-0.975849
H	-3.350020	-4.834771	0.451887
O	-2.394655	-4.038132	1.668947
H	-1.529122	-3.714776	1.325439
H	-2.197042	-4.606984	2.427175
O	-2.184249	5.104600	-0.601854
H	-2.739060	4.472646	-1.114674
H	-2.388911	5.988379	-0.941697
O	0.359868	4.360240	0.236614
H	-0.406682	4.680801	-0.288285
H	0.936749	5.130085	0.358449
O	-1.789406	4.445667	2.120635
H	-0.872341	4.226042	1.875533
H	-2.141571	4.825989	1.290832
O	6.294758	-2.353048	2.024743
H	5.528370	-1.959234	2.494178
H	6.952838	-2.571468	2.700693
O	2.418679	-2.228067	0.866862
H	2.956311	-1.944974	1.635431
H	2.189867	-1.401521	0.393426
O	0.493628	-2.549860	-2.062338
H	0.677269	-1.613029	-1.858438
H	1.390786	-2.962511	-2.038273
O	4.407403	-1.398235	-3.007613
H	4.695069	-0.674622	-2.421713
H	5.198081	-1.671851	-3.495465
O	3.064329	-3.405722	-1.601284
H	3.607256	-2.735386	-2.069384
H	3.042320	-3.122348	-0.664789
O	4.050519	-0.948295	2.733711
H	4.092428	-0.132549	2.191942
H	3.730046	-0.688634	3.610274
O	6.918521	-0.307194	0.383092
H	7.314290	0.384798	0.934317
H	6.745782	-1.077964	0.983584

Table S33. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-PPII_A'** conformation.

N	-4.567722	-1.009863	0.161536
C	-3.151659	-1.289554	-0.190727
C	-2.387354	0.014350	-0.016259
N	-1.076306	-0.074366	0.164955
C	-0.250936	1.110262	0.314250
C	1.045664	0.914476	-0.457966
N	1.736316	2.018618	-0.730576
C	3.066698	1.925472	-1.290627
C	3.981484	1.153973	-0.342481
O	5.029300	0.650451	-0.968298
O	-3.000048	1.094800	-0.109808
O	1.462703	-0.217821	-0.760486
C	3.638243	3.323020	-1.531375
O	3.785121	1.061039	0.855854
C	-3.048507	-1.788352	-1.629911
C	0.060661	1.379078	1.793019
H	-5.151316	-1.858166	-0.010169

H	-4.955149	-0.211787	-0.389467
H	-4.602497	-0.751275	1.160623
H	-2.784346	-2.041180	0.513307
H	-0.654879	-1.001815	0.307912
H	-0.801127	1.956631	-0.109231
H	1.342353	2.929664	-0.472825
H	3.028562	1.370160	-2.234223
H	5.722453	0.260629	-0.321329
H	3.699731	3.881182	-0.591872
H	2.995012	3.868537	-2.226730
H	4.637221	3.249452	-1.965458
H	-2.008375	-2.026991	-1.869121
H	-3.404315	-1.024958	-2.329800
H	-3.650132	-2.692601	-1.756954
H	0.664957	2.283995	1.903192
H	0.608715	0.534191	2.224003
H	-0.876764	1.509645	2.341763
O	-3.451785	-0.252403	2.623785
H	-3.436232	0.711896	2.787752
H	-3.484833	-0.681089	3.491219
O	-3.354952	2.507077	2.182795
H	-3.315082	2.160593	1.262933
H	-4.238201	2.895792	2.284712
O	-5.827621	1.173046	-1.177203
H	-6.455334	0.993712	-1.891766
H	-5.221236	1.872298	-1.511823
O	-3.798553	2.839782	-2.085944
H	-3.498836	3.685928	-1.688233
H	-3.247146	2.179595	-1.628115
O	-0.229688	-2.830249	0.431869
H	-0.033851	-2.930679	-0.533029
H	0.661074	-2.780910	0.842564
O	-5.886862	-3.365774	-0.554368
H	-6.697111	-3.627135	-0.093425
H	-5.253504	-4.119226	-0.457515
O	-3.964825	-5.254359	-0.272958
H	-3.477832	-5.452276	-1.085768
H	-3.306878	-4.888655	0.365736
O	-2.312608	-4.129583	1.576766
H	-1.468701	-3.773078	1.211880
H	-2.069814	-4.733291	2.293714
O	-2.473210	4.975800	-0.868440
H	-2.540025	5.050528	0.101540
H	-1.529460	4.764954	-0.998179
O	0.242316	4.403034	-0.100987
H	-0.245164	4.478260	0.750447
H	0.775464	5.210021	-0.172735
O	-1.667581	4.649420	1.855558
H	-2.209363	3.873624	2.128140
H	-1.568630	5.211016	2.638560
O	6.293223	-2.332402	2.091968
H	5.512255	-1.949806	2.546387
H	6.937413	-2.550657	2.781171
O	2.437548	-2.235257	0.843280
H	2.955371	-1.960063	1.627904
H	2.205637	-1.402411	0.382582
O	0.569410	-2.538230	-2.143741
H	0.733389	-1.599529	-1.933524
H	1.472082	-2.936841	-2.101552
O	4.460856	-1.307320	-3.005981
H	4.770537	-0.607078	-2.403181
H	5.232408	-1.563110	-3.532539
O	3.144290	-3.357371	-1.636072
H	3.681369	-2.671601	-2.088177

H	3.101993	-3.086361	-0.696623
O	4.017267	-0.957025	2.757309
H	4.062055	-0.137397	2.221608
H	3.677492	-0.705823	3.629063
O	6.933947	-0.269248	0.480225
H	7.315070	0.422289	1.042204
H	6.755744	-1.045124	1.072512

Table S34. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-PPII_B** conformation.

N	-4.599007	-0.939954	0.152609
C	-3.178071	-1.221012	-0.175256
C	-2.409710	0.077454	0.020447
N	-1.102349	-0.024556	0.204807
C	-0.253945	1.142745	0.368118
C	1.030417	0.934177	-0.423977
N	1.737338	2.026602	-0.694113
C	3.056559	1.910802	-1.275024
C	3.969329	1.108804	-0.351166
O	4.991821	0.582600	-1.000752
O	-3.016976	1.165765	-0.059875
O	1.421068	-0.205153	-0.740215
C	3.654209	3.299830	-1.503108
O	3.795821	1.012774	0.850597
C	-3.054231	-1.713253	-1.615418
C	0.081880	1.369174	1.848249
H	-5.176508	-1.794118	-0.014156
H	-4.975533	-0.161134	-0.430394
H	-4.650744	-0.665287	1.146215
H	-2.825382	-1.976965	0.531533
H	-0.692814	-0.959361	0.339175
H	-0.790995	2.011064	-0.026445
H	1.363964	2.949682	-0.425010
H	2.994525	1.369796	-2.225735
H	5.687570	0.173889	-0.369780
H	3.739773	3.841481	-0.556047
H	3.011270	3.869869	-2.178548
H	4.644884	3.212766	-1.953613
H	-2.012307	-1.957600	-1.840923
H	-3.396229	-0.945078	-2.317179
H	-3.659777	-2.613143	-1.755146
H	0.710730	2.255492	1.969502
H	0.614915	0.501711	2.252461
H	-0.844334	1.511167	2.412990
O	-3.479304	-0.248129	2.632831
H	-3.425793	0.706322	2.840134
H	-3.513950	-0.714360	3.480551
O	-3.308036	2.517893	2.302303
H	-3.302559	2.205530	1.370756
H	-4.182902	2.911436	2.447957
O	-5.749118	1.111067	-1.470207
H	-6.235734	0.822836	-2.256030
H	-5.120437	1.797487	-1.774870
O	-3.602402	2.883874	-2.052553
H	-3.265404	2.779466	-2.955592
H	-3.135019	2.220711	-1.503448
O	-0.283392	-2.785889	0.471690
H	-0.105368	-2.894689	-0.495572
H	0.615233	-2.746793	0.866518

O	-5.902036	-3.300188	-0.563410
H	-6.717243	-3.559721	-0.110176
H	-5.268975	-4.052457	-0.454944
O	-3.979102	-5.180686	-0.244547
H	-3.464821	-5.362962	-1.044112
H	-3.346264	-4.809877	0.416332
O	-2.385463	-4.030389	1.640901
H	-1.531108	-3.697146	1.278700
H	-2.163433	-4.618848	2.377065
O	-2.111429	5.020529	-0.871583
H	-2.255488	4.965558	0.092732
H	-2.700137	4.356938	-1.282319
O	0.465359	4.439190	-0.002401
H	0.039753	4.470818	0.876136
H	-0.293434	4.613041	-0.599409
O	-1.586554	4.646260	1.901809
H	-2.145991	3.900169	2.212758
H	-1.644938	5.335572	2.579724
O	6.259370	-2.439742	2.029008
H	5.495124	-2.041039	2.497719
H	6.913133	-2.667144	2.706156
O	2.400151	-2.242704	0.833301
H	2.934770	-1.982844	1.611942
H	2.179210	-1.403148	0.379139
O	0.475773	-2.504694	-2.117616
H	0.665660	-1.571856	-1.901956
H	1.369337	-2.925082	-2.091866
O	4.371514	-1.358845	-3.038284
H	4.690349	-0.661246	-2.437092
H	5.138910	-1.623567	-3.566505
O	3.036815	-3.385840	-1.654215
H	3.016028	-3.113826	-0.714311
H	3.581385	-2.711647	-2.114832
O	4.027625	-1.014473	2.738071
H	4.078717	-0.192696	2.206140
H	3.701288	-0.763168	3.614883
O	6.907136	-0.385629	0.405911
H	7.311541	0.296283	0.963370
H	6.725946	-1.159972	0.998999

Table S35. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-PPII_C** conformation.

N	-4.669429	-0.580806	0.015698
C	-3.266125	-1.000638	-0.226525
C	-2.380602	0.196503	0.090808
N	-1.154341	-0.065134	0.521190
C	-0.169786	0.972156	0.775370
C	0.983300	0.841110	-0.213474
N	1.612886	1.962737	-0.541841
C	2.864256	1.895779	-1.265466
C	3.875735	1.071892	-0.472241
O	4.819363	0.561612	-1.243515
O	-2.823204	1.346434	-0.106421
O	1.362293	-0.275545	-0.617676
C	3.417145	3.303584	-1.490556
O	3.840873	0.950374	0.739271
C	-3.090792	-1.425870	-1.682659
C	0.388206	0.844940	2.196665
H	-5.313408	-1.372841	-0.209550

H	-4.917687	0.236531	-0.582772
H	-4.764630	-0.320583	1.009685
H	-3.047110	-1.832192	0.450697
H	-0.887026	-1.046179	0.687027
H	-0.655686	1.943351	0.645422
H	1.253539	2.871878	-0.202852
H	2.711627	1.395018	-2.227626
H	5.576099	0.142038	-0.695769
H	3.594715	3.803983	-0.533818
H	2.698812	3.891690	-2.067442
H	4.355805	3.254579	-2.046167
H	-2.070969	-1.783059	-1.851230
H	-3.291251	-0.584285	-2.354060
H	-3.785848	-2.237004	-1.917369
H	1.137533	1.620400	2.377474
H	0.860899	-0.133145	2.339504
H	-0.424958	0.951192	2.920471
O	-3.814921	-0.018607	2.648068
H	-3.485792	0.895843	2.768710
H	-3.201815	-0.595735	3.126279
O	-2.990896	2.630486	2.319687
H	-2.989894	2.431197	1.359011
H	-3.742222	3.225180	2.471932
O	-5.424112	1.506697	-1.771833
H	-5.904087	1.212416	-2.559561
H	-4.685286	2.062892	-2.093940
O	-3.010924	2.902967	-2.310289
H	-2.591816	2.621367	-3.138034
H	-2.689176	2.291101	-1.615170
O	-0.505435	-2.852490	0.731990
H	-0.348252	-2.931851	-0.243291
H	0.400987	-2.871819	1.108074
O	-6.074480	-2.837141	-0.791004
H	-6.920288	-3.057640	-0.374679
H	-5.474743	-3.607692	-0.633579
O	-4.254235	-4.794822	-0.372169
H	-3.674583	-4.961478	-1.129418
H	-3.662065	-4.597368	0.392274
O	-2.620647	-4.237040	1.744659
H	-1.804564	-3.769684	1.450509
H	-2.998026	-3.708052	2.462398
O	-1.606874	5.138495	-1.355575
H	-2.171117	4.426482	-1.724868
H	-2.211891	5.788793	-0.970944
O	-0.665469	4.054294	2.857962
H	-0.152487	3.658702	3.576982
H	-1.481348	3.517268	2.767133
O	0.467717	4.353778	0.345931
H	0.104912	4.285758	1.255587
H	-0.281442	4.620533	-0.230204
O	6.235559	-2.592429	1.493012
H	5.531425	-2.199460	2.052441
H	6.930313	-2.897960	2.094259
O	2.209852	-2.398336	0.925660
H	2.853104	-2.131403	1.614981
H	1.988909	-1.569583	0.449219
O	0.211906	-2.553495	-1.865058
H	0.472233	-1.633760	-1.664726
H	1.078104	-3.027262	-1.898685
O	3.990128	-1.534932	-3.059430
H	4.337587	-0.789639	-2.536715
H	4.711329	-1.803764	-3.647148
O	2.753650	-3.551070	-1.573852
H	3.276785	-2.875282	-2.056138

H	2.776602	-3.284761	-0.632093
O	4.130185	-1.162108	2.524496
H	4.147730	-0.336356	1.995881
H	3.974147	-0.902723	3.444786
O	6.847805	-0.463742	-0.046068
H	7.295632	0.179415	0.524062
H	6.683734	-1.266401	0.513429

Table S36. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_β-PPII_D** conformation.[a]

N	-4.638978	-0.494174	-0.063584
C	-3.250695	-1.006548	-0.187879
C	-2.316233	0.157151	0.116904
N	-1.123431	-0.148652	0.615352
C	-0.106195	0.854657	0.875943
C	0.996407	0.758522	-0.171123
N	1.551895	1.897715	-0.573952
C	2.774252	1.858759	-1.349647
C	3.855047	1.128314	-0.554438
O	4.783325	0.601320	-1.331881
O	-2.683886	1.314898	-0.152912
O	1.412263	-0.347538	-0.565288
C	3.243906	3.276503	-1.674422
O	3.876800	1.087448	0.662563
C	-3.013618	-1.527619	-1.603400
C	0.514578	0.637185	2.258261
H	-5.309823	-1.268983	-0.259832
H	-4.811211	0.287668	-0.735332
H	-4.773829	-0.142987	0.900203
H	-3.124213	-1.804361	0.550517
H	-0.900070	-1.134258	0.809263
H	-0.581647	1.837747	0.825455
H	1.181134	2.795578	-0.242638
H	2.604984	1.297666	-2.274464
H	5.570014	0.232650	-0.787088
H	3.445621	3.836260	-0.755996
H	2.472244	3.797765	-2.246650
H	4.156377	3.240827	-2.272873
H	-2.008149	-1.947935	-1.691808
H	-3.124758	-0.716918	-2.331156
H	-3.739004	-2.312072	-1.836147
H	1.289803	1.384660	2.444722
H	0.961575	-0.360845	2.324948
H	-0.260068	0.734951	3.022391
O	-3.930503	0.282752	2.526010
H	-3.729078	1.238448	2.601679
H	-4.336328	0.018334	3.364355
O	-3.101230	2.893794	2.038078
H	-2.984649	2.574831	1.117233
H	-3.761148	3.603312	2.003868
O	-5.198549	1.574120	-1.922561
H	-5.642959	1.303338	-2.738779
H	-4.400111	2.084193	-2.197795
O	-2.782303	2.770632	-2.510513
H	-2.616623	3.653906	-2.112921
H	-2.414759	2.163468	-1.843208
O	-0.503427	-2.946441	0.822665
H	-0.319847	-3.012676	-0.148948

H	0.392242	-2.971720	1.220871
O	-6.103825	-2.765016	-0.746766
H	-6.934477	-2.964905	-0.291454
H	-5.513744	-3.545807	-0.607565
O	-4.331405	-4.785122	-0.399355
H	-3.793959	-4.990553	-1.177827
H	-3.699885	-4.649476	0.347274
O	-2.621648	-4.425143	1.697166
H	-1.814211	-3.919860	1.444989
H	-3.009475	-3.959507	2.452173
O	-2.039343	5.039103	-1.092803
H	-2.686014	5.346051	-0.441198
H	-1.239044	4.789886	-0.580405
O	0.274702	4.334298	0.326004
H	0.028300	4.193331	1.276851
H	0.873064	5.097400	0.311647
O	-0.563641	3.670608	2.797259
H	-1.512258	3.451775	2.660293
H	-0.539129	4.361358	3.476182
O	6.306694	-2.277083	1.631618
H	5.548701	-1.907315	2.134112
H	7.000147	-2.470317	2.279339
O	2.215382	-2.469685	1.004522
H	2.852583	-2.140990	1.671697
H	1.965622	-1.669362	0.493524
O	0.259885	-2.655675	-1.765402
H	0.517444	-1.729527	-1.594533
H	1.126330	-3.129442	-1.792888
O	3.992201	-1.635202	-2.996327
H	4.296032	-0.849962	-2.505729
H	4.729081	-1.886540	-3.572262
O	2.803425	-3.647567	-1.472639
H	3.317436	-2.969976	-1.962281
H	2.825674	-3.374556	-0.532935
O	4.043269	-0.992337	2.517979
H	4.047089	-0.187587	1.958089
H	3.850332	-0.702208	3.422113
O	6.862695	-0.315742	-0.135403
H	7.341392	0.381796	0.337558
H	6.709106	-1.043684	0.521356

This minimum has been obtained with the "Integral(UltraFineGrid)" option.

Table S37. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-PPII_A** conformation.

N	-5.272828	0.045713	0.279898
C	-4.008951	-0.665085	-0.043249
C	-2.895965	0.372421	0.010208
N	-1.673935	-0.073719	0.272270
C	-0.517567	0.803001	0.301459
C	0.564464	0.238708	-0.611966
N	1.434153	1.118444	-1.098917
C	2.609388	0.676763	-1.816353
C	3.448449	-0.244796	-0.940796
O	4.279995	-0.994859	-1.616030
O	-3.167545	1.562171	-0.235509
O	0.658718	-0.983371	-0.831573
C	3.452022	1.888174	-2.222333

O	3.385487	-0.239655	0.285858
C	-4.089995	-1.291476	-1.432999
C	0.035535	0.942034	1.724605
H	-6.084094	-0.603543	0.176759
H	-5.416164	0.868682	-0.342950
H	-5.204974	0.382698	1.255768
H	-3.859660	-1.426781	0.726473
H	-1.556594	-1.068037	0.508474
H	-0.835610	1.783226	-0.065787
H	1.326582	2.115869	-0.860169
H	2.313993	0.107483	-2.704574
H	4.973300	-1.460652	-0.964459
H	3.776114	2.444937	-1.336434
H	2.857754	2.551542	-2.856089
H	4.333548	1.565739	-2.779741
H	-3.167300	-1.835608	-1.654717
H	-4.235631	-0.518399	-2.194719
H	-4.926404	-1.994311	-1.478396
H	0.880302	1.636813	1.735282
H	0.372170	-0.028069	2.106295
H	-0.750847	1.327010	2.380241
O	-3.968899	0.584066	2.699603
H	-3.610105	1.501049	2.732552
H	-4.273436	0.371941	3.593509
O	-3.129281	3.097184	2.108648
H	-3.197497	2.795089	1.183473
H	-2.253168	3.541524	2.148464
O	-5.837685	2.351520	-1.333648
H	-6.504329	2.343176	-2.035245
H	-5.064398	2.829411	-1.694413
O	-3.295769	3.460092	-2.116147
H	-3.091737	3.299516	-3.050318
H	-3.006611	2.661210	-1.627428
O	-1.675602	-2.929652	0.718613
H	-1.562424	-3.137657	-0.242881
H	-0.792203	-3.118662	1.100575
O	-7.245748	-1.861782	-0.236378
H	-8.085235	-1.815905	0.243669
H	-6.869528	-2.758667	-0.057439
O	-5.990639	-4.213067	0.254963
H	-5.614715	-4.638159	-0.529181
H	-5.231943	-4.005212	0.851513
O	-4.016051	-3.473012	1.976580
H	-3.115699	-3.407296	1.580654
H	-3.934272	-4.046566	2.752313
O	-1.348801	5.133761	-1.013861
H	-1.351175	6.021898	-1.400309
H	-2.080882	4.640246	-1.447983
O	1.006382	3.775462	-0.277129
H	1.841782	4.297930	-0.354102
H	0.325833	4.254941	-0.793505
O	-0.827429	4.545115	1.706302
H	-0.024391	4.102899	1.369646
H	-1.181072	4.960659	0.894436
O	4.648914	-4.359856	0.533780
H	4.134346	-3.857766	1.202468
H	5.063832	-5.099271	1.001426
O	1.065157	-3.072949	0.942869
H	1.765443	-2.968492	1.614841
H	1.059232	-2.241645	0.423131
O	-0.978879	-3.042126	-1.900535
H	-0.530081	-2.183234	-1.783400
H	-0.231587	-3.687634	-1.873599
O	3.128908	-3.240587	-3.066882

H	3.545769	-2.459358	-2.661197
H	3.860335	-3.792286	-3.381137
O	1.268812	-4.550305	-1.442037
H	1.972866	-4.120046	-1.972844
H	1.364756	-4.197262	-0.534399
O	3.505749	-2.373487	2.081886
H	3.520604	-1.654191	1.415723
H	4.223591	-2.117791	2.690126
O	5.955393	-2.068228	-0.097324
H	5.980030	-1.587073	0.763345
H	5.633239	-2.980784	0.107926
O	4.377066	1.556829	2.078139
H	3.728457	1.114649	1.494247
H	3.885779	1.868081	2.854260
O	5.806871	-0.820587	2.385428
H	6.627545	-0.769667	2.897934
H	5.430944	0.087531	2.369016
O	3.375699	5.113185	-0.527772
H	4.128314	4.631435	-0.110099
H	3.377137	5.999531	-0.138498
O	5.413083	3.698047	0.623583
H	6.093690	3.367948	0.019346
H	5.099221	2.922551	1.136527

Table S38. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-PPII_B** conformation.

N	-5.267378	0.072972	0.547993
C	-4.048239	-0.597357	0.029807
C	-2.917398	0.417858	0.100876
N	-1.682836	-0.057997	0.081855
C	-0.525478	0.819735	0.094726
C	0.559793	0.209496	-0.780837
N	1.481526	1.054035	-1.237817
C	2.697882	0.569394	-1.852689
C	3.472550	-0.301379	-0.868925
O	4.440479	-0.983679	-1.427625
O	-3.189084	1.638184	0.103182
O	0.609399	-1.015428	-0.987228
C	3.566605	1.754724	-2.278068
O	3.234915	-0.317673	0.334185
C	-4.270171	-1.054779	-1.409938
C	0.013531	1.003158	1.520072
H	-6.086400	-0.572216	0.474807
H	-5.475051	0.937226	0.003996
H	-5.100909	0.326854	1.534513
H	-3.836783	-1.446007	0.685635
H	-1.539953	-1.075524	0.148718
H	-0.829763	1.790090	-0.310541
H	1.394451	2.053739	-1.015909
H	2.454874	-0.052679	-2.721561
H	5.070265	-1.409265	-0.689935
H	3.839934	2.362373	-1.408631
H	3.012931	2.381647	-2.981840
H	4.477912	1.400171	-2.762698
H	-3.385481	-1.585442	-1.771524
H	-4.458886	-0.196029	-2.063104
H	-5.126659	-1.732624	-1.460618
H	0.847682	1.710686	1.527658
H	0.360601	0.045888	1.923502

H	-0.782707	1.391313	2.161910
O	-3.657868	0.337708	2.835208
H	-3.312658	1.230268	3.038739
H	-3.706971	-0.138647	3.676615
O	-2.770218	2.952349	2.468078
H	-3.029751	2.672644	1.563870
H	-3.431942	3.602390	2.751173
O	-5.953810	2.338311	-1.060161
H	-6.617615	2.180878	-1.747285
H	-5.206906	2.791501	-1.500184
O	-3.436136	3.333362	-1.990790
H	-3.245366	2.966119	-2.867953
H	-3.169288	2.645475	-1.343002
O	-1.667841	-2.943444	0.336203
H	-1.487512	-3.182103	-0.607608
H	-0.809155	-3.107487	0.781721
O	-7.310364	-1.758370	0.058994
H	-8.055951	-1.797551	0.675385
H	-6.928914	-2.670502	0.025256
O	-6.045230	-4.151487	-0.017965
H	-5.739525	-4.430532	-0.892936
H	-5.237075	-4.028373	0.536010
O	-3.946207	-3.631383	1.630285
H	-3.071899	-3.518116	1.187936
H	-3.818146	-4.287390	2.330923
O	-1.360553	5.042032	-1.255445
H	-1.422180	5.943118	-1.605485
H	-2.137176	4.551628	-1.607215
O	1.060661	3.797653	-0.575673
H	0.667909	3.916277	0.320490
H	0.370520	4.189045	-1.150645
O	-0.644312	4.498917	1.419616
H	-1.266839	3.936328	1.921834
H	-1.152487	4.828498	0.651657
O	4.795050	-4.296572	0.925604
H	4.176162	-3.806707	1.510286
H	5.228694	-4.963392	1.477543
O	1.049834	-3.059532	0.822661
H	1.671607	-2.941689	1.566259
H	1.067378	-2.222711	0.313500
O	-0.768092	-3.186764	-2.215580
H	-0.397589	-2.290165	-2.121465
H	0.020852	-3.767457	-2.088182
O	3.348756	-3.057626	-3.107141
H	3.870993	-2.381868	-2.638570
H	3.988137	-3.590665	-3.601950
O	1.542890	-4.507908	-1.535516
H	2.239839	-4.024782	-2.028652
H	1.545577	-4.138646	-0.629298
O	3.347267	-2.340149	2.226811
H	3.409341	-1.650965	1.531120
H	3.957503	-2.005812	2.909519
O	5.977849	-1.927407	0.308201
H	5.873400	-1.411802	1.144981
H	5.700747	-2.852862	0.518245
O	3.983536	1.649189	2.190712
H	3.437645	1.210203	1.515002
H	4.460829	2.368297	1.713859
O	5.467084	-0.576102	2.661949
H	6.211585	-0.422344	3.262039
H	5.015669	0.299418	2.538438
O	3.553392	4.953279	-0.820921
H	2.637046	4.604413	-0.728834
H	3.498348	5.901125	-0.630887

O	5.291219	3.658358	0.850932
H	6.033103	3.345548	0.313732
H	4.691472	4.144477	0.236671

Table S39. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-PPII_C** conformation.

N	-5.156092	0.312436	0.347713
C	-3.922403	-0.473281	0.097552
C	-2.753281	0.499557	0.152193
N	-1.564073	-0.018644	0.428745
C	-0.334759	0.754464	0.367028
C	0.541840	0.211608	-0.753555
N	1.183386	1.099316	-1.509220
C	2.260941	0.644809	-2.366934
C	3.321089	-0.029297	-1.497504
O	4.046095	-0.925973	-2.108286
O	-2.948302	1.703628	-0.111248
O	0.702781	-1.014287	-0.901082
C	2.877644	1.822629	-3.121330
O	3.496463	0.291697	-0.322864
C	-3.994688	-1.142521	-1.273451
C	0.444631	0.655219	1.680723
H	-5.988703	-0.317490	0.299433
H	-5.261742	1.064531	-0.365906
H	-5.085480	0.737430	1.286475
H	-3.834354	-1.217984	0.893726
H	-1.510782	-1.015499	0.682863
H	-0.599218	1.794822	0.160888
H	1.060143	2.103400	-1.313702
H	1.881031	-0.099065	-3.073925
H	4.840876	-1.235726	-1.459380
H	3.290534	2.560616	-2.426059
H	2.112707	2.305624	-3.734674
H	3.678825	1.469991	-3.774147
H	-3.110787	-1.765211	-1.437773
H	-4.050482	-0.388259	-2.065703
H	-4.881626	-1.779861	-1.328584
H	1.377971	1.223658	1.611166
H	0.689658	-0.388435	1.905967
H	-0.157494	1.061273	2.498754
O	-3.873312	0.868862	2.771740
H	-3.355064	1.698185	2.826253
H	-4.170423	0.668773	3.671107
O	-2.375897	3.137540	2.165597
H	-2.587182	2.893371	1.239680
H	-2.856665	3.961375	2.342327
O	-5.508822	2.221472	-1.745613
H	-6.101358	1.957445	-2.464364
H	-4.681103	2.526681	-2.169849
O	-2.865313	2.925416	-2.547457
H	-2.580960	2.338827	-3.265660
H	-2.663078	2.448753	-1.714785
O	-1.640426	-2.833402	0.967949
H	-1.551971	-3.098928	0.016702
H	-0.754880	-3.028138	1.340882
O	-7.159456	-1.581155	-0.003137
H	-7.985704	-1.511256	0.496557
H	-6.786603	-2.476013	0.194269
O	-5.957214	-3.953359	0.488803

H	-5.612771	-4.389738	-0.303604
H	-5.194100	-3.858625	1.107821
O	-3.891660	-3.688640	2.251222
H	-3.041179	-3.407900	1.840491
H	-4.056677	-3.078026	2.984126
O	-0.897289	4.922239	-2.465682
H	-1.659043	4.305560	-2.520083
H	-1.265789	5.787325	-2.235722
O	0.269963	3.992906	1.903214
H	0.966785	3.551084	2.428276
H	-0.596590	3.615044	2.155323
O	0.917100	3.806952	-0.724161
H	0.578604	3.926684	0.197315
H	0.306915	4.289796	-1.329380
O	5.141316	-3.546443	1.097506
H	4.525719	-2.940907	1.567332
H	5.802409	-3.826982	1.746906
O	1.109171	-2.975410	0.985353
H	1.877077	-2.666510	1.509441
H	0.969791	-2.244295	0.344521
O	-1.037330	-3.160963	-1.654433
H	-0.569601	-2.305798	-1.688116
H	-0.299795	-3.818396	-1.624009
O	2.720436	-3.250632	-3.192605
H	3.205998	-2.460442	-2.892507
H	3.354152	-3.766091	-3.712580
O	1.245128	-4.633336	-1.271042
H	1.856459	-4.182384	-1.892413
H	1.397369	-4.220697	-0.396410
O	3.357164	-1.585048	1.799329
H	3.448372	-1.021221	1.003999
H	3.482517	-0.954511	2.539755
O	5.902191	-1.545249	-0.574500
H	5.961903	-0.817387	0.095385
H	5.711226	-2.364015	-0.052777
O	4.059810	0.613837	3.291774
H	4.418636	0.570990	4.190933
H	3.532849	1.447603	3.241353
O	6.103914	0.400258	1.341404
H	6.915097	0.312070	1.863260
H	5.380944	0.510096	1.995789
O	3.390725	3.049460	0.302801
H	3.395258	2.094563	0.088896
H	2.628516	3.431304	-0.179745
O	2.712022	2.981941	2.889685
H	3.111218	3.676181	3.436729
H	3.048941	3.123382	1.964013

Table S40. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_β-PPII_D** conformation.

N	5.230149	0.433048	-0.020591
C	4.041688	-0.455152	0.041793
C	2.815477	0.424541	-0.161174
N	1.741951	-0.151643	-0.690668
C	0.479341	0.552890	-0.835495
C	-0.467774	0.146986	0.287589
N	-1.231264	1.101116	0.809016
C	-2.307792	0.743797	1.708566
C	-3.245427	-0.238848	1.015380

O	-3.919383	-1.000955	1.834407
O	2.853972	1.610602	0.212913
O	-0.570739	-1.043328	0.642877
C	-3.089515	1.994188	2.113889
O	-3.391270	-0.262638	-0.204672
C	3.983889	-1.146474	1.402032
C	-0.167617	0.212545	-2.178519
H	6.091429	-0.143026	0.099710
H	5.191212	1.160469	0.728348
H	5.244600	0.902646	-0.943335
H	4.130040	-1.186226	-0.768270
H	1.775013	-1.146692	-0.950096
H	0.681549	1.625821	-0.779180
H	-1.114116	2.081116	0.510920
H	-1.903382	0.245830	2.596200
H	-4.696566	-1.507213	1.307787
H	-3.526659	2.482571	1.236936
H	-2.419975	2.700129	2.612794
H	-3.893227	1.727772	2.803599
H	3.142499	-1.843546	1.442281
H	3.868403	-0.404810	2.199541
H	4.907787	-1.706323	1.572717
H	-1.110274	0.754654	-2.288697
H	-0.365300	-0.862405	-2.251909
H	0.502031	0.510740	-2.988545
O	4.350025	1.279089	-2.535453
H	3.909441	2.153665	-2.506169
H	4.822960	1.234909	-3.379077
O	2.841012	3.487975	-1.775941
H	2.808738	3.038767	-0.904238
H	3.255218	4.351759	-1.627054
O	5.209944	2.379675	2.045906
H	5.709143	2.165742	2.847137
H	4.303815	2.632893	2.343662
O	2.564277	2.838115	2.680816
H	2.165663	3.679206	2.363460
H	2.366185	2.217956	1.956192
O	1.748227	-2.997689	-1.079578
H	1.655997	-3.173210	-0.107820
H	0.861510	-3.215599	-1.433533
O	7.248045	-1.444238	0.393641
H	8.089491	-1.373146	-0.079604
H	6.866964	-2.324790	0.155999
O	6.024779	-3.791126	-0.191917
H	5.612663	-4.219347	0.572185
H	5.334378	-3.754868	-0.896466
O	4.148011	-3.697808	-2.170407
H	3.244041	-3.483607	-1.842645
H	4.334815	-3.072273	-2.885210
O	1.230843	4.961144	1.499946
H	1.755359	5.512532	0.902069
H	0.522840	4.557663	0.944747
O	-0.775041	3.784207	0.010693
H	-0.535730	3.770643	-0.945957
H	-1.606941	4.311061	0.100059
O	0.178386	3.484850	-2.534711
H	1.147261	3.574355	-2.399758
H	-0.064249	4.146596	-3.199102
O	-4.907798	-4.358190	-0.589815
H	-4.428332	-3.851398	-1.278543
H	-5.561939	-4.902541	-1.051592
O	-1.020986	-3.203186	-1.022798
H	-1.841679	-3.057908	-1.537353
H	-0.923865	-2.393483	-0.476038

O	1.159235	-3.099891	1.563250
H	0.664826	-2.260660	1.494435
H	0.446741	-3.783301	1.603750
O	-2.530346	-3.173957	3.164423
H	-3.008115	-2.409855	2.794139
H	-3.168375	-3.632627	3.730323
O	-1.058978	-4.696807	1.350008
H	-1.673180	-4.215800	1.945084
H	-1.225963	-4.344788	0.451490
O	-3.525304	-2.420299	-1.981303
H	-3.631747	-1.643873	-1.391334
H	-3.732167	-2.115949	-2.877686
O	-5.794654	-2.091362	0.616939
H	-6.018288	-1.515248	-0.156074
H	-5.550643	-2.975495	0.246136
O	-5.258484	3.694754	-0.296135
H	-6.033386	4.151679	-0.653812
H	-5.043952	2.977195	-0.930680
O	-3.076089	5.187684	0.408782
H	-3.098980	6.058633	-0.013106
H	-3.889429	4.715697	0.111862
O	-4.501020	1.547844	-1.894407
H	-4.118128	1.755281	-2.760579
H	-3.813487	1.071452	-1.386169
O	-6.411954	-0.487027	-1.516026
H	-6.585766	-0.965403	-2.339632
H	-5.785905	0.232000	-1.744472

Table S41. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **PPII-β** conformation.

N	-4.280424	-1.322107	-0.961574
C	-3.785245	-0.288759	0.002465
C	-2.376414	0.069237	-0.474080
N	-1.907894	1.265468	-0.101130
C	-0.512493	1.607041	-0.311698
C	0.375901	0.573629	0.396814
N	1.558027	0.334819	-0.201759
C	2.531639	-0.565332	0.380496
C	3.889749	-0.220090	-0.196315
O	4.866741	-0.900401	0.405673
O	-1.725002	-0.750416	-1.119655
O	0.040479	0.051688	1.456329
C	2.189350	-2.037616	0.114019
O	4.071796	0.555411	-1.107954
C	-3.734880	-0.876256	1.409234
C	-0.233996	3.000614	0.246861
H	-5.062465	-1.864604	-0.580968
H	-3.503117	-1.961568	-1.186649
H	-4.592194	-0.907739	-1.846861
H	-4.464670	0.563460	-0.051738
H	-2.480502	1.879152	0.467123
H	-0.308197	1.581996	-1.387784
H	1.796975	0.776965	-1.083258
H	2.564971	-0.391422	1.461635
H	5.718190	-0.669426	-0.010527
H	2.198253	-2.241357	-0.960876
H	1.189522	-2.240290	0.504805
H	2.906295	-2.695543	0.610526

H	-3.335655	-0.130848	2.100277
H	-3.084406	-1.755406	1.436941
H	-4.736911	-1.153135	1.746161
H	0.807655	3.276193	0.067600
H	-0.419732	3.022941	1.325345
H	-0.875757	3.738314	-0.242420

Table S42. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 2H_2O$ peptide-water complex in the **$2H_2O$_PPII-β** conformation.

N	-3.906556	0.083735	-1.336780
C	-3.290732	-0.944408	-0.448628
C	-1.961013	-0.351261	0.004922
O	-1.335867	0.395979	-0.755588
N	-1.508816	-0.717953	1.205599
C	-0.166679	-0.357315	1.632578
C	0.843085	-0.849348	0.585477
O	0.705997	-1.931892	0.022836
N	1.899639	-0.040514	0.386668
C	2.961693	-0.398099	-0.531204
C	4.173524	0.445794	-0.190978
O	4.165196	1.380382	0.578520
O	5.251184	0.053786	-0.872138
C	-3.057570	-2.245813	-1.208979
C	0.135397	-1.000586	2.983600
C	2.554903	-0.185515	-1.995860
H	-3.262455	0.314497	-2.102630
H	-4.065890	0.964249	-0.784327
H	-4.791218	-0.239451	-1.738821
H	-3.972790	-1.088282	0.392371
H	-2.049752	-1.356072	1.778384
H	-0.107974	0.733582	1.717050
H	1.967571	0.856773	0.855682
H	3.220533	-1.451112	-0.375246
H	5.998785	0.639971	-0.650979
H	-4.002879	-2.643382	-1.585760
H	-2.614788	-2.986430	-0.539384
H	-2.374450	-2.081382	-2.047581
H	-0.586964	-0.665027	3.732584
H	1.135610	-0.714334	3.316731
H	0.091662	-2.091056	2.903485
H	1.659865	-0.776897	-2.201522
H	3.355236	-0.505674	-2.667056
H	2.332234	0.870132	-2.178051
O	-3.891932	2.232014	0.361293
H	-4.065442	2.047123	1.295915
H	-2.998882	2.648034	0.312879
O	-1.349402	3.105557	-0.135371
H	-1.004744	2.219715	-0.358787
H	-0.770197	3.460830	0.554682

Table S43. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_PPII-β_A** conformation.

N	-4.018179	0.265625	-0.172000

C	-3.083175	-0.307894	-1.177411
C	-1.765639	-0.572001	-0.462920
O	-1.390004	0.183577	0.447020
N	-1.042381	-1.603773	-0.892285
C	0.315335	-1.818285	-0.424523
C	1.175988	-0.589100	-0.760448
O	0.902427	0.162597	-1.690980
N	2.270489	-0.446229	0.010780
C	3.287824	0.543644	-0.285191
C	4.571480	0.094774	0.385556
O	4.654762	-0.839290	1.151491
O	5.600837	0.871636	0.047620
C	-2.862909	0.677362	-2.323188
C	0.895427	-3.062813	-1.095385
C	2.894125	1.950263	0.181005
H	-3.697283	1.213967	0.148154
H	-4.085556	-0.374630	0.653148
H	-4.952674	0.375170	-0.574953
H	-3.522532	-1.241048	-1.538192
H	-1.383834	-2.168706	-1.662430
H	0.291542	-1.955778	0.662119
H	2.471482	-1.106096	0.755916
H	3.458549	0.559102	-1.368106
H	6.399790	0.561674	0.513600
H	-3.813923	0.908684	-2.810351
H	-2.191341	0.239012	-3.064767
H	-2.416135	1.603784	-1.949027
H	0.279023	-3.935887	-0.865486
H	1.907611	-3.250783	-0.731432
H	0.935814	-2.927803	-2.180873
H	1.971276	2.241984	-0.324826
H	3.678208	2.668729	-0.069397
H	2.729112	1.959804	1.262857
O	-3.847180	-1.604346	1.872199
H	-3.809316	-2.530239	1.591533
H	-3.034049	-1.436149	2.402369
O	-3.263947	2.841378	0.541669
H	-3.705716	3.518281	0.008195
H	-2.294744	2.979511	0.422383
O	-0.556505	2.835629	0.178491
H	-0.056592	3.181703	0.932783
H	-0.527235	1.863103	0.259368
O	-1.550374	-0.734715	3.080074
H	-1.205485	-0.344228	2.254513
H	-0.866363	-1.339945	3.401804

Table S44. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_PPII-β_B** conformation.

N	-4.024726	0.240366	0.100967
C	-3.151713	-0.218259	-1.016416
C	-1.782561	-0.504630	-0.418457
O	-1.313327	0.257017	0.442404
N	-1.123979	-1.560511	-0.888343
C	0.255874	-1.820754	-0.510967
C	1.118160	-0.597604	-0.855452
O	0.870839	0.120692	-1.819599
N	2.187830	-0.422663	-0.056650
C	3.196889	0.576848	-0.346020
C	4.463086	0.178342	0.386345

O	4.537448	-0.726967	1.186775
O	5.488451	0.965409	0.058812
C	-3.032995	0.865276	-2.083783
C	0.769096	-3.043795	-1.268733
C	2.757584	1.989321	0.058189
H	-3.723258	1.180876	0.461666
H	-3.981730	-0.459018	0.872284
H	-4.995351	0.325929	-0.213533
H	-3.596520	-1.129739	-1.422327
H	-1.551569	-2.140398	-1.602217
H	0.292505	-2.004525	0.568851
H	2.360911	-1.047296	0.724749
H	3.408439	0.561167	-1.421610
H	6.276588	0.686230	0.561205
H	-4.018172	1.110092	-2.488800
H	-2.401761	0.508262	-2.900937
H	-2.583279	1.769229	-1.661597
H	0.156488	-3.917976	-1.032110
H	1.800358	-3.258836	-0.980138
H	0.738312	-2.864934	-2.348053
H	1.843178	2.240068	-0.483965
H	3.530965	2.717801	-0.196473
H	2.562311	2.033352	1.134170
O	-3.351038	-1.845975	1.814883
H	-3.442881	-2.753336	1.490299
H	-2.446640	-1.773301	2.196874
O	-3.327846	2.800303	0.920771
H	-3.888051	3.478075	0.514713
H	-2.400819	3.008653	0.657122
O	-0.713605	2.951141	0.143221
H	-0.126228	3.348262	0.803314
H	-0.627117	1.983552	0.246537
O	-0.772385	-1.321660	2.642438
H	-0.712926	-0.522952	2.084201
H	-0.656448	-1.027734	3.558088

Table S45. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_PPII-β_A** conformation.

N	2.821557	-1.630236	0.202567
C	2.259953	-0.278956	-0.044178
C	0.859942	-0.233118	0.548598
O	0.136450	-1.243332	0.545700
N	0.445569	0.948900	0.999019
C	-0.929193	1.149766	1.413736
C	-1.874954	0.768076	0.271765
O	-1.563151	0.975224	-0.912769
N	-3.059861	0.277985	0.633477
C	-4.131845	0.028914	-0.313703
C	-5.444854	0.287081	0.403076
O	-5.549793	0.448607	1.597646
O	-6.473671	0.274901	-0.443198
C	2.192886	0.004982	-1.542972
C	-1.138837	2.619331	1.781342
C	-4.100736	-1.401026	-0.867196
H	2.325505	-2.357368	-0.359510
H	2.739479	-1.848742	1.213348
H	3.832112	-1.644032	-0.065941
H	2.909575	0.444858	0.458283
H	1.070544	1.758766	0.933340

H	-1.141881	0.513023	2.279685
H	-3.273392	0.140206	1.617406
H	-4.037434	0.748649	-1.133319
H	-7.300403	0.408471	0.057269
H	3.190139	-0.077742	-1.983070
H	1.824975	1.019915	-1.716713
H	1.516528	-0.702736	-2.034642
H	-0.451498	2.900227	2.583675
H	-2.161691	2.782438	2.127519
H	-0.954118	3.262250	0.914416
H	-3.131705	-1.593229	-1.333862
H	-4.886515	-1.534158	-1.614349
H	-4.246237	-2.125321	-0.060005
O	2.279753	-1.597516	2.967323
H	2.393791	-0.749710	3.420384
H	1.358358	-1.890365	3.148650
O	1.658317	-3.526521	-1.528030
H	2.166276	-3.594925	-2.349436
H	0.752107	-3.225463	-1.781929
O	-0.838085	-2.514152	-1.761533
H	-1.060086	-1.843619	-2.443884
H	-0.688571	-2.005105	-0.941984
O	-0.307981	-2.522006	2.976773
H	-0.480378	-2.113065	2.107202
H	-1.002823	-2.205152	3.572054
O	-1.593633	-0.364043	-3.342698
H	-2.437168	-0.447250	-3.811294
H	-1.762428	0.218051	-2.575493
O	0.261777	2.882587	-1.783000
H	-0.385474	2.212224	-1.471495
H	-0.259245	3.631058	-2.109606
O	2.099260	3.129258	0.156980
H	1.447942	3.250821	-0.579021
H	2.132560	3.964493	0.647990
O	6.201062	0.939910	0.655620
H	5.581520	1.530957	0.165606
H	5.969636	1.015545	1.592459
O	5.536401	-1.471952	-0.408102
H	5.789592	-1.462431	-1.342462
H	5.854025	-0.617616	-0.025219
O	4.483718	2.301249	-0.947795
H	3.666212	2.688211	-0.562459
H	4.891745	3.003306	-1.475180

Table S46. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_PPII-β_B** conformation.

N	-2.615285	-1.770459	-0.625488
C	-2.277351	-0.425516	-0.088148
C	-0.828289	-0.143332	-0.451787
O	0.008164	-1.064261	-0.452861
N	-0.518390	1.119875	-0.722918
C	0.828243	1.510708	-1.101729
C	1.826293	0.993167	-0.068154
O	1.606917	1.119696	1.148653
N	2.953419	0.481192	-0.559761
C	4.038009	0.016416	0.282862
C	5.332371	0.187633	-0.489914
O	5.394723	0.423426	-1.674715

O	6.395773	0.002222	0.292594
C	-2.470655	-0.386595	1.424667
C	0.916004	3.034383	-1.172815
C	3.857036	-1.451250	0.690640
H	-2.190513	-2.528006	-0.047535
H	-2.271839	-1.844424	-1.599274
H	-3.654144	-1.896675	-0.611635
H	-2.934779	0.296589	-0.581822
H	-1.246796	1.837323	-0.670991
H	1.062586	1.071273	-2.078476
H	3.040754	0.336574	-1.563215
H	4.075657	0.650785	1.173951
H	7.205866	0.081137	-0.245301
H	-3.504818	-0.636964	1.676523
H	-2.254251	0.617330	1.800045
H	-1.795664	-1.096157	1.914582
H	0.197833	3.414497	-1.904294
H	1.918812	3.337740	-1.481904
H	0.698265	3.477508	-0.195799
H	2.906143	-1.569451	1.215709
H	4.667433	-1.766609	1.352158
H	3.846395	-2.092223	-0.196516
O	-1.350632	-1.069658	-3.058831
H	-1.723909	-1.050310	-3.951922
H	-0.378842	-1.152258	-3.164858
O	-1.692073	-3.771709	1.146743
H	-2.347025	-3.936444	1.840573
H	-0.875559	-3.451946	1.597983
O	0.657809	-2.614611	1.791135
H	0.917105	-1.983324	2.496302
H	0.544639	-2.061945	0.993565
O	1.458779	-1.190932	-2.771982
H	1.234526	-1.395178	-1.843009
H	1.935571	-1.960869	-3.116542
O	1.590724	-0.534374	3.389901
H	2.445562	-0.701354	3.813450
H	1.765659	0.097926	2.664073
O	-0.587334	2.477481	2.212246
H	0.176564	2.000754	1.817314
H	-0.209735	3.164002	2.781728
O	-2.397281	3.011560	0.290623
H	-1.766542	3.007478	1.053954
H	-2.436246	3.922132	-0.039571
O	-6.183823	0.703168	-0.962128
H	-5.697595	1.238840	-0.290493
H	-5.932013	1.055216	-1.827839
O	-5.391802	-1.861101	-0.564148
H	-5.784639	-2.161906	0.267974
H	-5.733741	-0.945483	-0.717061
O	-4.836492	1.964814	1.032667
H	-3.995380	2.414372	0.792274
H	-5.361121	2.616342	1.520166

Table S47. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_PPII-β_A** conformation.

N	3.094724	0.925453	1.569866
C	2.589752	0.310459	0.318255
C	1.102849	0.614613	0.200653

O	0.395120	0.719504	1.215792
N	0.612743	0.697334	-1.036061
C	-0.817363	0.766214	-1.273161
C	-1.503324	-0.471115	-0.683351
O	-0.894675	-1.551065	-0.569993
N	-2.784175	-0.326248	-0.355158
C	-3.565384	-1.444889	0.141714
C	-5.023400	-1.197754	-0.183793
O	-5.474613	-0.131353	-0.546928
O	-5.763093	-2.282853	0.018812
C	2.800050	-1.201457	0.341689
C	-1.081009	0.846996	-2.776346
C	-3.400152	-1.631756	1.656751
H	2.715568	0.442711	2.414470
H	2.831895	1.932853	1.590157
H	4.137054	0.853152	1.598117
H	3.137817	0.762193	-0.514819
H	1.240445	0.538721	-1.829986
H	-1.223639	1.655018	-0.778402
H	-3.240314	0.581382	-0.503448
H	-3.243481	-2.352911	-0.378394
H	-6.699666	-2.070773	-0.155479
H	3.855685	-1.432707	0.507539
H	2.507770	-1.632451	-0.618408
H	2.197779	-1.664817	1.130966
H	-0.583764	1.727212	-3.193027
H	-2.152759	0.928661	-2.968356
H	-0.704833	-0.047269	-3.284929
H	-2.343330	-1.776590	1.893536
H	-3.961779	-2.505263	1.995970
H	-3.756522	-0.743622	2.187717
O	2.208862	3.549846	1.172356
H	2.410498	3.896017	0.291078
H	1.225140	3.596426	1.273606
O	2.299093	-0.558478	3.829185
H	2.956512	-1.239851	4.031703
H	1.463597	-1.033759	3.595543
O	-0.059064	-1.493106	2.910663
H	-0.044047	-2.324916	2.381575
H	-0.093834	-0.770986	2.254620
O	-0.475517	3.394694	1.526687
H	-0.490433	2.420416	1.496654
H	-0.938674	3.703590	0.714754
O	-0.251447	-3.629154	1.203211
H	0.591567	-3.919886	0.790237
H	-0.676243	-3.072157	0.524010
O	0.991537	-2.577262	-2.260160
H	0.296093	-2.164546	-1.692786
H	0.517950	-3.070048	-2.949029
O	2.405433	-0.368005	-3.026643
H	1.943228	-1.236390	-2.961839
H	2.333192	-0.079933	-3.949760
O	6.346062	1.457036	-1.152944
H	5.831953	0.747428	-1.605228
H	5.946250	2.297925	-1.417296
O	5.882017	0.815007	1.449305
H	6.304734	-0.039311	1.618583
H	6.127590	1.068897	0.526282
O	4.984287	-0.699907	-2.091882
H	4.103482	-0.593388	-2.513400
H	5.497966	-1.268946	-2.683425
O	-5.717727	2.350833	0.754678
H	-6.073778	1.493755	0.462610
H	-6.453660	2.979664	0.720632

O	-3.700649	2.377545	-1.063226
H	-4.433692	2.508779	-0.412197
H	-4.140623	2.301142	-1.923884
O	-1.737510	4.269489	-0.759136
H	-2.479241	3.662747	-0.980138
H	-1.188622	4.336229	-1.553954
O	2.035534	-4.269378	-0.265563
H	2.101167	-5.205770	-0.503527
H	1.783642	-3.799425	-1.087352

Table S48. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_PPII-β_B** conformation.

N	3.160150	0.900547	1.539126
C	2.628828	0.321834	0.280293
C	1.137523	0.616296	0.215091
O	0.463254	0.676510	1.259249
N	0.608626	0.738229	-0.998923
C	-0.828947	0.816194	-1.189698
C	-1.497222	-0.441056	-0.622327
O	-0.876480	-1.519756	-0.544676
N	-2.771364	-0.310099	-0.271096
C	-3.533517	-1.446018	0.213794
C	-4.995908	-1.221951	-0.110074
O	-5.453127	-0.185900	-0.543021
O	-5.730826	-2.295777	0.165888
C	2.852817	-1.187848	0.249331
C	-1.143854	0.954584	-2.678117
C	-3.351208	-1.658670	1.723005
H	2.813158	0.379468	2.375697
H	2.873663	1.896348	1.607597
H	4.204687	0.851296	1.534483
H	3.149861	0.806538	-0.551374
H	1.212419	0.613906	-1.817292
H	-1.213203	1.688963	-0.652587
H	-3.232835	0.604921	-0.407066
H	-3.202526	-2.342182	-0.322466
H	-6.667303	-2.102417	-0.028437
H	3.915219	-1.414163	0.373354
H	2.533288	-1.590547	-0.714368
H	2.279822	-1.681131	1.042155
H	-0.640168	1.837866	-3.080786
H	-2.220175	1.074621	-2.815699
H	-0.807769	0.070300	-3.230827
H	-2.289910	-1.794913	1.945227
H	-3.898152	-2.545212	2.051868
H	-3.712823	-0.785736	2.274860
O	2.114964	3.504974	1.248332
H	2.182298	3.897256	0.365629
H	1.166162	3.541059	1.491792
O	2.420054	-0.657216	3.763151
H	3.083452	-1.338668	3.944921
H	1.581594	-1.132243	3.539212
O	0.040537	-1.584520	2.892836
H	0.030507	-2.404711	2.345735
H	-0.012749	-0.849629	2.252689
O	-0.585960	3.139949	1.831358
H	-0.461575	2.169796	1.767908
H	-0.989068	3.321128	2.693869

O	-0.224241	-3.665472	1.129074
H	0.608434	-3.946117	0.689231
H	-0.652617	-3.072103	0.483072
O	0.953470	-2.497437	-2.317545
H	0.276067	-2.098713	-1.718326
H	0.457728	-2.963815	-3.009139
O	2.344257	-0.266305	-3.067687
H	1.883530	-1.135967	-3.006834
H	2.241750	0.041165	-3.981600
O	6.328791	1.542516	-1.265376
H	5.805478	0.842005	-1.721344
H	5.924240	2.388641	-1.504528
O	5.940491	0.846106	1.334188
H	6.377953	-0.006205	1.473459
H	6.156589	1.121568	0.409737
O	4.951642	-0.599172	-2.215892
H	4.059553	-0.491756	-2.612831
H	5.454198	-1.153914	-2.830025
O	-6.293265	2.447355	-0.029351
H	-6.378098	1.494547	-0.207666
H	-6.970479	2.881261	-0.568065
O	-3.668263	2.286809	-0.986097
H	-4.553218	2.524243	-0.641402
H	-3.067882	3.043701	-0.820687
O	-1.600669	4.079602	-0.572161
H	-1.272158	3.901867	0.334191
H	-1.732740	5.036864	-0.632751
O	2.032294	-4.259194	-0.407774
H	2.091045	-5.186162	-0.681819
H	1.764420	-3.758532	-1.206264

Table S49. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}18H_2O$ peptide-water complex in the **$18H_2O$_PPII-β_A** conformation.

N	-4.409239	0.770682	-1.306198
C	-3.654283	0.276820	-0.129036
C	-2.191410	0.652717	-0.310144
O	-1.693708	0.717498	-1.448887
N	-1.480205	0.834928	0.800028
C	-0.033919	0.947803	0.743763
C	0.564562	-0.329478	0.141737
O	-0.033311	-1.419882	0.225475
N	1.763442	-0.200206	-0.412755
C	2.516040	-1.351098	-0.882075
C	3.994840	-1.036209	-0.749380
O	4.405526	0.111208	-0.643784
O	4.747190	-2.106036	-0.797294
C	-3.779529	-1.240626	-0.011988
C	0.524471	1.172492	2.148399
C	2.177569	-1.712197	-2.333335
H	-4.147084	0.250912	-2.173765
H	-4.217925	1.782314	-1.428534
H	-5.434678	0.641669	-1.144925
H	-4.067241	0.767799	0.758016
H	-1.936743	0.700567	1.707573
H	0.234244	1.791923	0.099477
H	2.218454	0.721432	-0.411640
H	2.289913	-2.200319	-0.230002
H	5.692850	-1.900029	-0.462470
H	-4.832906	-1.528715	0.041926

H	-3.288016	-1.582187	0.901687
H	-3.311655	-1.736628	-0.869680
H	0.086845	2.075231	2.583027
H	1.608882	1.297283	2.103899
H	0.297808	0.319883	2.797571
H	1.106780	-1.914115	-2.421372
H	2.732795	-2.601914	-2.640931
H	2.431041	-0.881773	-2.999264
O	-3.528263	3.429807	-1.057601
H	-3.580432	3.838878	-0.181783
H	-2.612449	3.569763	-1.376849
O	-3.897574	-0.833901	-3.559655
H	-4.548379	-1.549900	-3.595784
H	-3.015607	-1.262810	-3.430011
O	-1.384638	-1.639739	-2.988507
H	-1.273664	-2.435901	-2.417086
H	-1.256367	-0.874967	-2.396345
O	-0.915544	3.302022	-1.998667
H	-0.970590	2.323216	-2.005039
H	-0.737836	3.580374	-2.910247
O	-0.818276	-3.668082	-1.233073
H	-1.566556	-3.959034	-0.666362
H	-0.319579	-3.042924	-0.673439
O	-1.502097	-2.388212	2.307526
H	-0.939860	-1.995683	1.596366
H	-0.890976	-2.806674	2.934308
O	-2.848455	-0.163515	3.135601
H	-2.366271	-1.020149	3.056181
H	-2.634073	0.196976	4.010032
O	-7.171109	1.297835	1.913849
H	-6.543698	0.652886	2.317704
H	-6.796946	2.177673	2.064963
O	-7.111497	0.512380	-0.685970
H	-7.491556	-0.377075	-0.725708
H	-7.208011	0.813574	0.250899
O	-5.534250	-0.702911	2.750101
H	-4.597488	-0.514649	2.977914
H	-5.890096	-1.222144	3.485884
O	6.574693	1.744058	-0.177794
H	5.919034	1.082395	-0.489775
H	6.556400	2.467184	-0.822873
O	2.755498	2.498446	-0.064950
H	3.338082	2.670828	-0.820879
H	2.103681	3.241358	-0.039858
O	0.796977	4.372749	-0.093188
H	0.313219	4.558197	0.724785
H	0.134063	4.069454	-0.749838
O	4.663629	1.898174	1.906448
H	3.921246	2.107358	1.300732
H	5.458637	2.044872	1.357095
O	8.808633	0.158608	-0.052493
H	8.165170	0.899815	-0.083259
H	9.409982	0.348442	0.682015
O	6.883219	-1.718810	0.523725
H	7.620014	-1.099383	0.315413
H	6.358864	-1.326380	1.259773
O	4.873791	-0.821355	2.144973
H	4.872385	-1.022946	3.092003
H	4.735619	0.155982	2.075512
O	-2.791717	-4.268466	0.651009
H	-2.762950	-5.183759	0.966249
H	-2.424164	-3.722366	1.376791

Table S50. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_PPII-β_B** conformation.

N	-4.316822	0.765110	-1.515183
C	-3.692755	0.300587	-0.251902
C	-2.203880	0.605739	-0.317120
O	-1.607201	0.593814	-1.409435
N	-1.585761	0.818074	0.841781
C	-0.137015	0.892694	0.908825
C	0.476035	-0.418904	0.406723
O	-0.144461	-1.496383	0.512237
N	1.706125	-0.331100	-0.080445
C	2.466586	-1.514004	-0.444016
C	3.938947	-1.222250	-0.217818
O	4.357927	-0.080226	-0.071181
O	4.688286	-2.292900	-0.231158
C	-3.896708	-1.202492	-0.076785
C	0.301803	1.155000	2.348311
C	2.234952	-1.922293	-1.904538
H	-4.024749	0.171585	-2.323636
H	-4.047528	1.752316	-1.690118
H	-5.357962	0.713007	-1.431716
H	-4.159191	0.851969	0.570603
H	-2.124782	0.749689	1.710624
H	0.211615	1.708017	0.267491
H	2.168811	0.592265	-0.105574
H	2.170230	-2.334505	0.217176
H	5.664176	-2.045343	0.016838
H	-4.961695	-1.445817	-0.121845
H	-3.521375	-1.515385	0.899743
H	-3.361854	-1.759833	-0.853836
H	-0.162546	2.073578	2.717673
H	1.386959	1.273599	2.386962
H	0.011704	0.325048	3.001826
H	1.170100	-2.103949	-2.071738
H	2.790779	-2.834030	-2.136884
H	2.559845	-1.122176	-2.576870
O	-3.324771	3.405022	-1.477609
H	-3.464308	3.887197	-0.649836
H	-2.365401	3.460669	-1.670764
O	-3.726153	-1.013786	-3.614965
H	-4.387362	-1.720844	-3.634781
H	-2.860079	-1.445440	-3.407485
O	-1.267607	-1.821531	-2.848271
H	-1.211540	-2.587735	-2.229600
H	-1.160612	-1.029190	-2.288927
O	-0.594977	3.090334	-1.937435
H	-0.698118	2.115761	-1.912127
H	-0.194562	3.310566	-2.792370
O	-0.875729	-3.755145	-0.946816
H	-1.677998	-4.006575	-0.438472
H	-0.419152	-3.109320	-0.373847
O	-1.849088	-2.305965	2.471571
H	-1.204702	-1.962607	1.805147
H	-1.318355	-2.714548	3.173517
O	-3.189129	-0.011227	3.091679
H	-2.733289	-0.886111	3.084236
H	-3.038505	0.374250	3.968800
O	-7.345794	1.567504	1.450772
H	-6.774570	0.913924	1.919011
H	-6.965787	2.440394	1.625410

O	-7.076492	0.727407	-1.121326
H	-7.518967	-0.131244	-1.185449
H	-7.243947	1.056561	-0.204282
O	-5.844146	-0.462647	2.456752
H	-4.928140	-0.300525	2.772366
H	-6.279062	-0.981331	3.149083
O	6.347086	1.504936	-1.089082
H	5.673647	0.819012	-0.885173
H	6.075956	1.908975	-1.927755
O	2.656857	2.341970	0.068765
H	3.503702	2.424892	0.550562
H	2.089536	3.101273	0.311073
O	0.666133	4.260446	0.268066
H	0.041357	4.404362	0.993149
H	0.140546	3.936000	-0.492127
O	5.249882	2.500707	1.217025
H	5.533372	3.310341	1.667716
H	5.780574	2.433200	0.393672
O	8.519798	-0.198657	-1.157165
H	7.898637	0.558834	-1.214377
H	9.339588	0.147958	-0.776177
O	7.004172	-1.671139	0.604277
H	7.600585	-1.216577	-0.035597
H	6.761713	-0.998242	1.286837
O	6.056828	0.105536	2.446347
H	6.659044	0.417943	3.137394
H	5.696175	0.914802	2.022428
O	-3.022134	-4.229510	0.779263
H	-3.038644	-5.125671	1.145926
H	-2.710768	-3.648514	1.504453

Table S51. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_PPII-β_A** conformation.

N	4.936403	-0.647193	-1.346812
C	4.113826	-0.282756	-0.167275
C	2.703631	-0.806612	-0.399261
O	2.249003	-0.903141	-1.553307
N	1.987926	-1.083254	0.687871
C	0.570450	-1.383823	0.602145
C	-0.178220	-0.228788	-0.072410
O	0.286045	0.925471	-0.071035
N	-1.362954	-0.539863	-0.588694
C	-2.276140	0.477981	-1.078283
C	-3.692228	-0.044526	-0.905558
O	-3.929053	-1.213735	-0.646126
O	-4.612878	0.878234	-1.071600
C	4.088036	1.234151	0.007848
C	0.014018	-1.603587	2.009769
C	-2.012837	0.859552	-2.539488
H	4.611062	-0.140292	-2.200310
H	4.873866	-1.670623	-1.498806
H	5.933488	-0.392153	-1.164987
H	4.558912	-0.765325	0.709233
H	2.407000	-0.945077	1.614096
H	0.425729	-2.287938	0.000537
H	-1.696649	-1.509173	-0.516890
H	-2.170341	1.368246	-0.444954
H	-5.514472	0.578177	-0.646373
H	5.109813	1.613677	0.095480

H	3.545249	1.502037	0.917852
H	3.598149	1.709964	-0.848652
H	0.557013	-2.415204	2.502024
H	-1.043011	-1.872875	1.959861
H	0.115447	-0.691339	2.607727
H	-0.980465	1.203952	-2.648275
H	-2.684055	1.664443	-2.850643
H	-2.162745	-0.006847	-3.190679
O	4.358892	-3.397444	-1.174207
H	4.459007	-3.817302	-0.307676
H	3.463071	-3.631430	-1.494752
O	4.173298	0.945644	-3.539832
H	4.735603	1.729754	-3.620497
H	3.253402	1.274873	-3.395340
O	1.571913	1.468801	-2.966629
H	1.270056	2.247205	-2.451851
H	1.519314	0.712000	-2.353798
O	1.732382	-3.551393	-2.089133
H	1.686401	-2.571858	-2.100235
H	1.558796	-3.852242	-2.994283
O	0.366070	3.365842	-1.281473
H	-0.444483	3.688673	-1.750415
H	0.100283	2.559476	-0.793164
O	1.548786	1.849735	2.238822
H	1.246796	1.492271	1.378620
H	0.755699	2.330379	2.579509
O	3.047129	-0.115424	3.170154
H	2.435195	0.659382	3.041764
H	2.805581	-0.536864	4.008821
O	7.531404	-0.837259	2.018696
H	6.814947	-0.302607	2.437089
H	7.281663	-1.765593	2.131062
O	7.548478	-0.059561	-0.582533
H	7.855756	0.857168	-0.628039
H	7.615168	-0.331069	0.365990
O	5.620073	0.836890	2.981297
H	4.710032	0.494986	3.133779
H	5.879301	1.284278	3.800001
O	-5.840235	-3.112016	0.003514
H	-5.292906	-2.411129	-0.409886
H	-5.728977	-3.902003	-0.547405
O	-1.966097	-3.326985	-0.069713
H	-2.541590	-3.637670	-0.785800
H	-1.204952	-3.956599	-0.031067
O	0.278154	-4.852729	-0.107549
H	0.817133	-4.942304	0.691959
H	0.857599	-4.461567	-0.795684
O	-3.817891	-2.736386	1.952181
H	-3.101549	-2.928477	1.309702
H	-4.622881	-3.058815	1.500612
O	-8.244488	-1.790496	-0.066835
H	-7.532506	-2.464416	-0.013425
H	-8.853861	-1.976603	0.662167
O	-6.588832	0.330334	0.363608
H	-7.240405	-0.394934	0.202408
H	-6.034750	0.072187	1.133014
O	-4.481433	-0.124407	2.116776
H	-4.720541	-0.068167	3.055899
H	-4.139737	-1.052892	1.995946
O	-1.903476	4.306685	-2.431348
H	-1.874815	5.272079	-2.502156
H	-2.605887	4.100244	-1.767209
O	-3.804015	3.563752	-0.638400
H	-3.511527	3.244206	0.240507

H	-4.280661	2.798568	-1.004939
O	0.260182	5.119054	0.844545
H	0.420901	4.550385	0.056773
H	1.130269	5.430535	1.132843
O	-0.689689	3.314440	2.729976
H	-0.458677	4.011623	2.076015
H	-1.494006	2.874797	2.389954
O	-3.203452	2.336476	1.831798
H	-3.538642	1.410451	1.869611
H	-3.767622	2.824931	2.452667

Table S52. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_PPII-β_B** conformation.

N	4.927064	-0.628887	-1.330217
C	4.090427	-0.276675	-0.156727
C	2.686656	-0.809498	-0.404872
O	2.242007	-0.895122	-1.564056
N	1.963411	-1.105182	0.671681
C	0.546247	-1.403922	0.564587
C	-0.191222	-0.240031	-0.104954
O	0.261244	0.918799	-0.048559
N	-1.353468	-0.547231	-0.670848
C	-2.270597	0.481826	-1.127523
C	-3.687450	-0.008336	-0.880874
O	-3.930764	-1.146063	-0.514725
O	-4.607283	0.908412	-1.093076
C	4.048941	1.239127	0.024376
C	-0.031346	-1.636072	1.961595
C	-2.060020	0.861555	-2.597014
H	4.607463	-0.118846	-2.183972
H	4.871877	-1.651799	-1.490186
H	5.920781	-0.368950	-1.137559
H	4.531215	-0.759180	0.721943
H	2.371844	-0.972283	1.603250
H	0.406370	-2.302477	-0.044916
H	-1.670224	-1.530538	-0.648193
H	-2.121936	1.371079	-0.500875
H	-5.496581	0.628408	-0.633200
H	5.066087	1.628221	0.123016
H	3.494549	1.497692	0.930150
H	3.562533	1.713447	-0.835040
H	0.501546	-2.455037	2.453226
H	-1.089154	-1.900841	1.891415
H	0.065192	-0.731473	2.572136
H	-1.029085	1.196965	-2.742525
H	-2.734705	1.672682	-2.882833
H	-2.241488	-0.002309	-3.243323
O	4.368658	-3.386770	-1.203155
H	4.461627	-3.810325	-0.337610
H	3.473255	-3.615999	-1.529120
O	4.182214	0.966086	-3.528727
H	4.734510	1.758422	-3.597272
H	3.254706	1.281610	-3.402086
O	1.563710	1.451343	-3.003840
H	1.261567	2.223309	-2.479190
H	1.518529	0.688015	-2.398278
O	1.741314	-3.544619	-2.109486
H	1.685562	-2.565804	-2.121410
H	1.549634	-3.849599	-3.009471

O	0.383934	3.336674	-1.295206
H	-0.426697	3.670300	-1.756183
H	0.114298	2.532158	-0.804923
O	1.504608	1.824117	2.267239
H	1.190976	1.469982	1.409129
H	0.725099	2.323912	2.610578
O	2.999249	-0.151977	3.173745
H	2.385511	0.622870	3.052477
H	2.755213	-0.584489	4.005978
O	7.504764	-0.802290	2.056589
H	6.776296	-0.280034	2.469907
H	7.268850	-1.734651	2.165281
O	7.530818	-0.026964	-0.545439
H	7.832776	0.891486	-0.591694
H	7.593579	-0.295540	0.404162
O	5.559613	0.836856	3.011487
H	4.654062	0.478747	3.153468
H	5.804377	1.281833	3.835905
O	-5.739230	-3.157394	-0.062765
H	-5.212096	-2.434966	-0.465426
H	-5.628749	-3.929854	-0.638318
O	-1.970119	-3.311373	-0.309309
H	-2.546866	-3.238807	0.476831
H	-1.232550	-3.913529	-0.079954
O	0.344485	-4.834375	-0.060464
H	0.907721	-4.901152	0.724038
H	0.903605	-4.458807	-0.771976
O	-3.772137	-2.834766	1.831756
H	-3.721318	-3.306898	2.676849
H	-4.581408	-3.154947	1.375604
O	-8.139014	-1.800952	-0.082856
H	-7.432047	-2.480180	-0.044352
H	-8.761296	-2.012097	0.628158
O	-6.557988	0.369631	0.400549
H	-7.181488	-0.375632	0.220385
H	-5.994105	0.100843	1.158297
O	-4.443485	-0.143481	2.154013
H	-4.658798	-0.166233	3.100131
H	-4.090351	-1.041654	1.950051
O	-1.883268	4.314177	-2.418680
H	-1.840277	5.279628	-2.480922
H	-2.579278	4.112565	-1.746606
O	-3.776934	3.577997	-0.615786
H	-3.494726	3.255707	0.265310
H	-4.256138	2.817407	-0.988860
O	0.290419	5.090656	0.833300
H	0.447332	4.516113	0.049081
H	1.163118	5.393409	1.122962
O	-0.700777	3.343695	2.753806
H	-0.456582	4.020750	2.083861
H	-1.512637	2.910449	2.423648
O	-3.217250	2.355995	1.869964
H	-3.529780	1.423630	1.914768
H	-3.800469	2.838212	2.477821

Table S53. M062X/6-31+G*/PCM optimized Cartesian coordinates of the Ala_3H^+ peptide in the **PPII–PPII** conformation.

N	4.507075	-1.111530	0.561532
C	3.726072	-0.230659	-0.363410

C	2.439988	0.114010	0.389612
O	2.034769	-0.633434	1.277674
N	1.791199	1.214331	-0.011802
C	0.455979	1.495659	0.480994
C	-0.483604	0.342246	0.104505
O	-0.322243	-0.327398	-0.910306
N	-1.523435	0.150090	0.943497
C	-2.575981	-0.779696	0.603974
C	-3.289819	-0.326964	-0.665939
O	-3.303807	0.801396	-1.096983
O	-3.949413	-1.345033	-1.231814
C	3.399178	-0.989716	-1.645473
C	-0.057845	2.797466	-0.131105
C	-3.581885	-0.890790	1.750078
H	3.862387	-1.784090	1.002004
H	4.946564	-0.581363	1.322019
H	5.247864	-1.624803	0.073123
H	4.333546	0.654247	-0.563186
H	2.144223	1.752497	-0.795420
H	0.501660	1.581510	1.572211
H	-1.651974	0.764698	1.738583
H	-2.134512	-1.760577	0.399968
H	-4.432584	-1.012957	-2.011134
H	4.313694	-1.256412	-2.180506
H	2.793672	-0.356082	-2.296859
H	2.829893	-1.896518	-1.420170
H	0.616051	3.621440	0.118028
H	-1.050452	3.032260	0.259704
H	-0.127012	2.706113	-1.219857
H	-3.072496	-1.233932	2.654026
H	-4.363824	-1.609218	1.496254
H	-4.047520	0.079369	1.950636

Table S54. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 2H_2O$ peptide-water complex in the **$2H_2O$_PPII-PPII** conformation.

N	-4.016837	-0.580619	-1.176767
C	-3.102632	-1.387919	-0.318520
C	-1.919520	-0.477632	-0.005144
O	-1.586418	0.394531	-0.814302
N	-1.268276	-0.703935	1.137793
C	-0.041020	0.007903	1.448228
C	0.986478	-0.227554	0.334710
O	1.068982	-1.296995	-0.259209
N	1.820815	0.806140	0.094239
C	2.948280	0.645568	-0.794307
C	3.935950	-0.363819	-0.216571
O	3.996527	-0.700221	0.942133
O	4.770641	-0.810360	-1.162861
C	-2.642816	-2.644103	-1.051829
C	0.523774	-0.491120	2.776818
C	3.647620	1.986216	-1.022034
H	-3.517147	-0.248262	-2.010059
H	-4.339058	0.270673	-0.645950
H	-4.829811	-1.124099	-1.479480
H	-3.657064	-1.638027	0.588804
H	-1.573243	-1.445392	1.758548
H	-0.265196	1.078495	1.515310

H	1.761323	1.645711	0.658491
H	2.592272	0.244615	-1.748735
H	5.416500	-1.415841	-0.753898
H	-3.502210	-3.263258	-1.321112
H	-1.988670	-3.230879	-0.403252
H	-2.089412	-2.381250	-1.958204
H	-0.206501	-0.341544	3.576636
H	1.431188	0.062032	3.028950
H	0.771769	-1.554909	2.709138
H	2.940300	2.700754	-1.450043
H	4.481245	1.861680	-1.715691
H	4.032107	2.388019	-0.079154
O	-4.472654	1.622222	0.384573
H	-4.575391	1.451063	1.332178
H	-3.699118	2.226805	0.285599
O	-2.220695	3.039993	-0.233517
H	-1.691160	2.252818	-0.464508
H	-1.710380	3.531089	0.426939

Table S55. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 4H_2O$ peptide-water complex in the **$4H_2O$_PPII-PPII** conformation.

N	3.004674	-1.569128	-0.517781
C	2.400374	-0.258013	-0.162510
C	1.059375	-0.555138	0.494052
O	0.504691	-1.648627	0.306864
N	0.496597	0.416438	1.212184
C	-0.878901	0.285049	1.666785
C	-1.774972	0.001888	0.454753
O	-1.577624	0.537788	-0.630573
N	-2.798342	-0.852132	0.668935
C	-3.815283	-1.034520	-0.342841
C	-4.590429	0.262433	-0.548862
O	-4.605498	1.199873	0.213303
O	-5.290607	0.223388	-1.689099
C	2.196730	0.587207	-1.419310
C	-1.329793	1.575942	2.344537
C	-4.764383	-2.166318	0.050777
H	2.406032	-2.073103	-1.217206
H	3.107002	-2.165527	0.338372
H	3.931376	-1.436173	-0.931396
H	3.072917	0.237375	0.542548
H	0.962805	1.311150	1.319183
H	-0.938297	-0.552119	2.370952
H	-2.973015	-1.204376	1.603318
H	-3.319524	-1.278226	-1.288272
H	-5.799780	1.050479	-1.776164
H	3.152683	0.747773	-1.924611
H	1.782282	1.561754	-1.153351
H	1.505690	0.088104	-2.105402
H	-0.694578	1.791897	3.207658
H	-2.361125	1.473905	2.689822
H	-1.279334	2.412029	1.640394
H	-4.195902	-3.091550	0.176039
H	-5.511917	-2.318486	-0.730050
H	-5.278788	-1.936594	0.989595
O	3.119513	-2.947354	1.888518
H	3.489112	-2.439197	2.625124
H	2.218368	-3.232048	2.167649
O	1.436969	-2.809443	-2.465628

H	1.574011	-2.506546	-3.374944
H	0.468300	-2.758757	-2.295177
O	-1.209826	-2.600562	-1.687284
H	-1.586033	-3.465062	-1.462996
H	-0.926856	-2.209149	-0.839918
O	0.491296	-3.630148	2.270215
H	0.186213	-2.946971	1.643735
H	0.059682	-3.434415	3.114941

Table S56. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_PPII-PPII_A** conformation.

N	2.810303	-1.595318	-0.328008
C	2.226750	-0.243499	-0.148311
C	0.907368	-0.405235	0.588987
O	0.234252	-1.439639	0.447098
N	0.484676	0.633061	1.307534
C	-0.869065	0.651734	1.833747
C	-1.848380	0.404974	0.684998
O	-1.685853	0.954518	-0.416716
N	-2.893390	-0.386655	0.932930
C	-3.958521	-0.557890	-0.034989
C	-4.664017	0.774933	-0.269575
O	-4.588449	1.742477	0.448997
O	-5.415127	0.721481	-1.373869
C	1.972564	0.408320	-1.506526
C	-1.162968	2.009363	2.469482
C	-4.955685	-1.609003	0.452427
H	2.208228	-2.180520	-0.949563
H	2.890924	-2.050472	0.601117
H	3.761164	-1.515623	-0.754766
H	2.929581	0.350399	0.444401
H	1.057506	1.482248	1.357149
H	-0.977453	-0.142689	2.580157
H	-2.992519	-0.811340	1.848363
H	-3.528103	-0.880308	-0.989583
H	-5.888133	1.568131	-1.477926
H	2.906583	0.469331	-2.071624
H	1.584967	1.421887	-1.373082
H	1.240467	-0.176097	-2.075259
H	-0.457253	2.198393	3.282611
H	-2.176467	2.020457	2.877012
H	-1.074526	2.808920	1.726895
H	-4.440832	-2.561726	0.599781
H	-5.740747	-1.749036	-0.292424
H	-5.415548	-1.300294	1.396345
O	2.682528	-2.271400	2.400506
H	2.871951	-1.566055	3.035797
H	1.792546	-2.623452	2.628133
O	1.261752	-3.090493	-2.159567
H	1.543493	-2.971147	-3.078003
H	0.306415	-2.844114	-2.126444
O	-1.325467	-2.302861	-1.706882
H	-1.600872	-1.548505	-2.272403
H	-1.010229	-1.901894	-0.875741
O	0.115276	-3.250419	2.558977
H	-0.179471	-2.662970	1.837471
H	-0.470113	-3.076177	3.310460
O	-2.205748	-0.003042	-2.985761
H	-1.655758	0.362602	-3.694019

H	-2.048590	0.555254	-2.199755
O	-0.084906	3.166025	-0.924700
H	-0.664422	2.395087	-0.736966
H	-0.671446	3.932617	-1.007298
O	1.931704	3.056584	0.844310
H	1.201066	3.310917	0.226219
H	1.975766	3.741795	1.528666
O	6.139173	1.011348	0.112341
H	5.447668	1.687937	-0.081197
H	6.112263	0.856903	1.067473
O	5.384075	-1.174974	-1.310371
H	5.482739	-0.977543	-2.252811
H	5.726757	-0.385889	-0.822565
O	4.213998	2.760211	-0.681523
H	3.432888	2.937678	-0.111564
H	4.562446	3.626970	-0.936411

Table S57. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 10H_2O$ peptide-water complex in the **$10H_2O$_PPII-PPII_B** conformation.

N	3.012008	-1.545476	-0.555802
C	2.379977	-0.262438	-0.165384
C	1.056286	-0.586200	0.510667
O	0.515913	-1.691015	0.331755
N	0.491994	0.380997	1.230309
C	-0.878955	0.230542	1.685319
C	-1.770449	-0.068654	0.476628
O	-1.558787	0.437535	-0.628826
N	-2.816692	-0.882586	0.688168
C	-3.820151	-1.051149	-0.341033
C	-4.490635	0.286780	-0.629694
O	-4.439892	1.262929	0.088495
O	-5.172144	0.247748	-1.772038
C	2.140501	0.601788	-1.403202
C	-1.352427	1.516526	2.361242
C	-4.853702	-2.094132	0.081104
H	2.421149	-2.045744	-1.254864
H	3.130562	-2.149849	0.284054
H	3.948921	-1.346488	-0.969067
H	3.052707	0.241397	0.536254
H	0.940553	1.301844	1.297624
H	-0.934839	-0.605953	2.390920
H	-2.997997	-1.244369	1.617756
H	-3.319826	-1.372077	-1.261147
H	-5.613600	1.106303	-1.913922
H	3.086897	0.782115	-1.920728
H	1.720203	1.568326	-1.115391
H	1.443627	0.103833	-2.085369
H	-0.705313	1.752714	3.210347
H	-2.375217	1.393672	2.725575
H	-1.333542	2.351400	1.653464
H	-4.354799	-3.051883	0.249888
H	-5.599118	-2.223690	-0.705555
H	-5.362249	-1.790454	1.001449
O	3.169417	-2.878700	1.921101
H	3.499084	-2.324017	2.642645
H	2.279523	-3.194521	2.198281
O	1.401984	-2.781488	-2.537831
H	1.499377	-2.443460	-3.439697

H	0.439831	-2.758908	-2.334642
O	-1.221938	-2.647146	-1.632203
H	-1.563521	-3.521938	-1.393103
H	-0.878661	-2.263114	-0.803173
O	0.554074	-3.655172	2.304005
H	0.245008	-2.980453	1.669692
H	0.110742	-3.460588	3.142810
O	-3.068032	3.777619	-0.060732
H	-3.583324	2.953931	0.026854
H	-3.158064	4.236819	0.787024
O	-0.512926	2.996645	-0.831858
H	-0.732726	2.040066	-0.842878
H	-1.365032	3.415635	-0.577599
O	1.462401	3.072223	0.948700
H	0.700998	3.220716	0.322589
H	1.340875	3.679803	1.694173
O	5.746930	1.606017	0.085500
H	5.042028	2.279707	-0.069256
H	5.758544	1.436353	1.038207
O	5.509136	-0.646721	-1.413705
H	5.645088	-0.426451	-2.346358
H	5.648957	0.189203	-0.905401
O	3.807775	3.438809	-0.458487
H	2.986918	3.372473	0.079931
H	4.111089	4.354486	-0.375010

Table S58. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_PPII-PPII_A** conformation.

N	-3.419447	-0.530767	-1.486136
C	-2.758099	-0.342168	-0.172252
C	-1.406369	0.307542	-0.421380
O	-0.795544	0.105790	-1.484155
N	-0.893302	1.024884	0.576346
C	0.487727	1.468434	0.523493
C	1.406755	0.256505	0.368749
O	1.191051	-0.804619	0.980038
N	2.479423	0.416511	-0.407683
C	3.491493	-0.617358	-0.480966
C	4.077658	-0.847926	0.907218
O	4.212502	0.007667	1.755518
O	4.482374	-2.106412	1.080123
C	-2.555585	-1.689190	0.519036
C	0.837624	2.222668	1.804298
C	4.606753	-0.202966	-1.441144
H	-2.887112	-1.207820	-2.076542
H	-3.473692	0.388641	-1.966600
H	-4.387871	-0.896864	-1.344380
H	-3.393282	0.313428	0.431814
H	-1.424780	1.134638	1.446601
H	0.623505	2.129670	-0.337842
H	2.647442	1.322508	-0.856861
H	3.042132	-1.555119	-0.825878
H	4.910104	-2.192942	1.954044
H	-3.517906	-2.194498	0.636667
H	-2.120937	-1.543608	1.511557
H	-1.881777	-2.320569	-0.070604
H	0.161924	3.074527	1.920370
H	1.863375	2.597547	1.756344
H	0.741193	1.568154	2.677526

H	4.185476	-0.040299	-2.436234
H	5.358349	-0.992747	-1.506445
H	5.089943	0.718790	-1.101898
O	-3.270264	2.172714	-2.222991
H	-3.560436	2.795531	-1.541188
H	-2.349212	2.435198	-2.468257
O	-2.074434	-2.546688	-2.940039
H	-2.429999	-3.424836	-2.740312
H	-1.117360	-2.571689	-2.694094
O	0.531951	-2.251352	-2.188475
H	0.778399	-2.762712	-1.383029
H	0.317845	-1.354452	-1.871276
O	-0.653732	2.574855	-2.868329
H	-0.395556	1.703500	-2.515017
H	-0.231494	3.241898	-2.281145
O	1.383284	-3.502210	0.115003
H	2.289837	-3.747158	-0.168618
H	1.471640	-2.672279	0.619234
O	3.900374	-3.885445	-1.012115
H	4.163289	-4.742957	-1.378422
H	4.571303	-3.654507	-0.349818
O	-0.401446	-1.127223	3.223093
H	0.170016	-1.007068	2.432341
H	0.197407	-1.194899	3.981428
O	-2.290398	0.771415	3.065514
H	-1.602703	0.110710	3.333540
H	-2.261626	1.490488	3.714870
O	-6.568559	0.435245	1.093711
H	-5.889928	0.195058	1.768218
H	-6.386594	1.350048	0.834408
O	-6.034634	-1.317170	-0.915393
H	-6.190700	-2.215527	-0.590443
H	-6.311715	-0.705163	-0.190115
O	-4.705782	-0.536059	2.820065
H	-3.872087	-0.043303	2.989975
H	-5.062228	-0.769525	3.689588
O	3.026059	3.167439	-1.037797
H	3.632373	3.379935	-1.763698
H	3.557371	3.196557	-0.204236
O	4.195396	2.813594	1.375726
H	4.214465	1.846194	1.526070
H	5.096130	3.128737	1.542846
O	0.544849	4.349646	-1.102091
H	0.485533	5.284972	-1.345897
H	1.501131	4.126661	-1.077869

Table S59. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 14H_2O$ peptide-water complex in the **$14H_2O$_PPII-PPII_B** conformation.

N	-3.434373	-0.502636	-1.501237
C	-2.777867	-0.318836	-0.183814
C	-1.421859	0.322235	-0.427005
O	-0.811609	0.115466	-1.491741
N	-0.904971	1.034204	0.569424
C	0.477158	1.475773	0.513650
C	1.394750	0.263311	0.350109
O	1.157942	-0.813376	0.928914
N	2.485495	0.442247	-0.392504
C	3.500956	-0.589123	-0.448884

C	4.007752	-0.872270	0.960835
O	4.125987	-0.041200	1.835527
O	4.361186	-2.147990	1.124077
C	-2.588883	-1.668232	0.506734
C	0.835683	2.224602	1.795013
C	4.666505	-0.133386	-1.327182
H	-2.913371	-1.197152	-2.082594
H	-3.461910	0.409483	-1.992167
H	-4.411963	-0.845699	-1.360964
H	-3.411330	0.340999	0.417405
H	-1.436103	1.147030	1.439804
H	0.610560	2.140965	-0.345232
H	2.682588	1.376316	-0.788912
H	3.074746	-1.514012	-0.852946
H	4.734677	-2.269456	2.018489
H	-3.555135	-2.167904	0.615919
H	-2.159996	-1.527143	1.502295
H	-1.915518	-2.303028	-0.079532
H	0.166470	3.080795	1.916980
H	1.863858	2.592279	1.741489
H	0.740032	1.568669	2.667340
H	4.302006	0.060313	-2.339030
H	5.431079	-0.912056	-1.375048
H	5.114073	0.782859	-0.929454
O	-3.146063	2.202142	-2.247181
H	-3.451830	2.900653	-1.651006
H	-2.244914	2.456949	-2.535234
O	-2.112779	-2.542849	-2.932057
H	-2.470747	-3.417901	-2.723186
H	-1.153399	-2.569885	-2.693927
O	0.499458	-2.249776	-2.216533
H	0.777331	-2.765425	-1.423923
H	0.297248	-1.355259	-1.885994
O	-0.435277	2.465093	-2.863469
H	-0.299740	1.557736	-2.518141
H	-0.130476	2.461873	-3.783709
O	1.425557	-3.502574	0.054213
H	2.335803	-3.723506	-0.237280
H	1.496792	-2.671527	0.559683
O	3.953865	-3.841782	-1.069861
H	4.226367	-4.688532	-1.453773
H	4.604701	-3.633577	-0.380682
O	-0.405960	-1.131246	3.186972
H	0.152783	-1.015080	2.385986
H	0.205947	-1.200252	3.934701
O	-2.282445	0.780815	3.065346
H	-1.593608	0.115831	3.320333
H	-2.241010	1.497151	3.717091
O	-6.571918	0.428473	1.163350
H	-5.890825	0.183919	1.834047
H	-6.411746	1.355344	0.934613
O	-6.051177	-1.249126	-0.909214
H	-6.212753	-2.157765	-0.617209
H	-6.316269	-0.662581	-0.158589
O	-4.699351	-0.534104	2.882519
H	-3.863508	-0.037520	3.029152
H	-5.043879	-0.746737	3.762109
O	3.120020	3.099874	-1.038333
H	2.324778	3.674806	-1.045286
H	3.543591	3.203563	-0.160244
O	4.235086	2.778169	1.484006
H	4.202849	1.810485	1.623645
H	5.161671	3.032350	1.605581
O	0.654538	4.367166	-1.144804

H	0.598387	5.290628	-1.430941
H	0.216986	3.834467	-1.842018

Table S60. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_PPII-PPII_A** conformation.

N	4.453224	0.459479	0.901523
C	3.419283	-0.273284	0.132715
C	2.219016	0.646447	-0.024510
O	1.973330	1.517192	0.825799
N	1.421470	0.412490	-1.066380
C	0.106335	1.022213	-1.121079
C	-0.683960	0.637746	0.128320
O	-0.553182	-0.480179	0.654834
N	-1.569968	1.525233	0.587580
C	-2.536346	1.100290	1.586680
C	-3.291174	-0.109828	1.032289
O	-3.623739	-0.185835	-0.143881
O	-3.569410	-1.038043	1.914515
C	3.000629	-1.539278	0.878987
C	-0.634873	0.535407	-2.364982
C	-3.511165	2.230347	1.907288
H	4.114299	0.679047	1.865119
H	4.672357	1.336664	0.396250
H	5.317762	-0.123113	0.970340
H	3.842906	-0.519003	-0.846383
H	1.634143	-0.366023	-1.699819
H	0.207957	2.109851	-1.157693
H	-1.699764	2.407185	0.083937
H	-2.008985	0.794130	2.496478
H	-4.048668	-1.833794	1.428425
H	3.870164	-2.182561	1.040203
H	2.267782	-2.098343	0.290651
H	2.554893	-1.282296	1.846123
H	-0.045243	0.770494	-3.255130
H	-1.602629	1.034008	-2.452253
H	-0.801507	-0.546592	-2.317341
H	-2.960630	3.094445	2.287808
H	-4.220553	1.906066	2.672382
H	-4.071599	2.531076	1.016154
O	4.455044	2.494613	-1.030194
H	4.591449	2.243766	-1.954789
H	3.661151	3.081522	-1.009759
O	3.580674	0.823737	3.559649
H	3.854636	0.100645	4.142221
H	2.593492	0.862548	3.606758
O	0.869295	1.053942	3.367057
H	0.411147	0.182012	3.414619
H	0.967479	1.245678	2.416451
O	2.176899	3.948686	-0.650053
H	1.838577	3.308816	0.002932
H	1.554422	3.924921	-1.410913
O	-0.495837	-1.333149	3.351734
H	-1.319581	-1.156656	3.855311
H	-0.737977	-1.255193	2.410402
O	-2.869242	-0.541931	4.579525
H	-3.193814	-0.953184	5.394057
H	-3.476796	-0.816641	3.870610
O	-0.143862	-2.779173	-0.825358

H	-0.198632	-1.986677	-0.250986
H	-1.063048	-2.908432	-1.144535
O	1.829583	-2.071232	-2.456667
H	1.032045	-2.443318	-1.989689
H	1.646620	-2.110900	-3.407703
O	6.413755	-2.282867	-1.661592
H	5.531633	-2.722651	-1.717296
H	6.422119	-1.602545	-2.350050
O	6.680633	-1.213752	0.817698
H	6.731311	-1.927850	1.469195
H	6.637682	-1.646759	-0.070391
O	4.004545	-3.543818	-1.623533
H	3.228909	-3.059271	-1.986800
H	4.016102	-4.404024	-2.067869
O	-2.003054	3.566894	-1.443372
H	-2.389154	4.419081	-1.187669
H	-2.736920	3.035375	-1.827810
O	-3.868369	1.637778	-2.122399
H	-3.797126	1.126029	-1.281711
H	-4.768373	2.001034	-2.142699
O	0.406738	3.768079	-2.796775
H	0.494360	4.494351	-3.431504
H	-0.515772	3.804100	-2.462953
O	-4.629920	-2.911073	0.589267
H	-5.317061	-2.448738	0.047799
H	-3.922683	-3.094682	-0.069952
O	-4.175080	-1.001558	-2.985256
H	-4.327167	-1.215003	-3.918552
H	-3.990147	-0.038966	-2.941470
O	-6.243600	-1.470279	-1.111263
H	-5.570896	-1.288953	-1.802157
H	-6.359865	-0.623005	-0.654945
O	-2.842251	-2.980950	-1.551503
H	-3.196464	-2.198437	-2.033008
H	-2.997486	-3.736683	-2.139562

Table S61. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 18H_2O$ peptide-water complex in the **$18H_2O$_PPII-PPII_B** conformation.

N	-4.459171	-0.701883	0.728359
C	-3.466610	0.231438	0.142631
C	-2.236855	-0.578618	-0.230245
O	-1.980862	-1.639981	0.366646
N	-1.431607	-0.060900	-1.152615
C	-0.119260	-0.638156	-1.375731
C	0.671305	-0.631361	-0.067974
O	0.536818	0.284983	0.762309
N	1.561089	-1.610252	0.106160
C	2.541282	-1.479735	1.169970
C	3.282229	-0.155148	0.976402
O	3.597760	0.258360	-0.131891
O	3.570391	0.481131	2.086611
C	-3.087776	1.306708	1.159658
C	0.633007	0.178754	-2.425487
C	3.528703	-2.643486	1.126510
H	-4.104803	-1.110560	1.622778
H	-4.637995	-1.461826	0.049252
H	-5.348382	-0.186914	0.917622
H	-3.915272	0.675649	-0.751521

H	-1.654101	0.851254	-1.566282
H	-0.228059	-1.670303	-1.723499
H	1.712259	-2.292748	-0.652678
H	2.032546	-1.455442	2.139962
H	4.047802	1.381592	1.846768
H	-3.979637	1.854808	1.476010
H	-2.390968	2.019608	0.711025
H	-2.611493	0.852251	2.035307
H	0.058595	0.201072	-3.355843
H	1.606396	-0.273143	-2.629226
H	0.789188	1.205889	-2.077140
H	2.993437	-3.585957	1.266259
H	4.268241	-2.540764	1.924167
H	4.050974	-2.674147	0.164499
O	-4.232527	-2.178214	-1.635791
H	-4.922534	-2.409309	-2.274235
H	-3.574638	-2.903501	-1.660551
O	-3.537864	-1.634464	3.221164
H	-3.771795	-1.041231	3.949700
H	-2.551129	-1.707685	3.227002
O	-0.838184	-1.870671	2.921926
H	-0.356541	-1.060896	3.213450
H	-0.936573	-1.781233	1.956471
O	-1.970135	-3.788368	-1.343026
H	-1.762130	-3.200533	-0.586545
H	-1.985981	-4.693264	-0.995840
O	0.588502	0.383640	3.589489
H	1.408279	0.055316	4.017521
H	0.828263	0.565249	2.661927
O	2.952518	-0.761918	4.520680
H	3.304378	-0.578858	5.404253
H	3.543206	-0.312200	3.891278
O	0.102978	2.902832	-0.012531
H	0.149460	1.981139	0.319427
H	1.024363	3.107287	-0.281901
O	-1.839994	2.704866	-1.814770
H	-1.053587	2.919573	-1.241720
H	-1.629531	3.009629	-2.710530
O	-6.415636	2.689820	-0.973824
H	-5.539602	3.142202	-0.920828
H	-6.450220	2.268075	-1.844371
O	-6.725840	0.887927	1.028389
H	-6.829337	1.357674	1.868385
H	-6.662263	1.580768	0.325531
O	-4.024652	3.960022	-0.690215
H	-3.247450	3.557206	-1.139603
H	-4.059069	4.880175	-0.989989
O	2.087465	-3.128726	-2.225740
H	1.292934	-3.185163	-2.800717
H	2.681677	-2.460693	-2.618818
O	3.861123	-0.941659	-2.554606
H	3.783686	-0.707172	-1.601228
H	4.753175	-1.305468	-2.671467
O	-0.351851	-3.196099	-3.530878
H	-0.461617	-3.767209	-4.305108
H	-0.974501	-3.529686	-2.850220
O	6.232748	1.759186	-0.693430
H	6.361025	0.819399	-0.493921
H	5.551175	1.772713	-1.399654
O	4.626266	2.659081	1.349499
H	5.312252	2.371529	0.697002
H	3.912137	3.013228	0.772358
O	2.804807	3.286865	-0.666728
H	3.162152	2.660601	-1.337352

H	2.953428	4.170302	-1.039240
O	4.156961	1.820449	-2.608541
H	3.989436	0.881203	-2.842182
H	4.294087	2.296890	-3.441430

Table S62. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_PPII-PPII_A** conformation.

N	4.762688	0.982895	0.283242
C	3.947072	-0.208695	-0.062675
C	2.541424	0.286016	-0.352375
O	2.157751	1.368771	0.128602
N	1.749542	-0.492761	-1.079365
C	0.362409	-0.114181	-1.277807
C	-0.324389	0.088363	0.068830
O	-0.030799	-0.600628	1.060393
N	-1.302517	0.999133	0.093172
C	-2.279367	0.942770	1.164119
C	-2.861204	-0.468410	1.202303
O	-2.971490	-1.148257	0.194367
O	-3.264481	-0.858482	2.396384
C	3.932665	-1.199101	1.099850
C	-0.384830	-1.188019	-2.065713
C	-3.403059	1.944036	0.903797
H	4.427041	1.413743	1.172851
H	4.675406	1.684944	-0.473549
H	5.759902	0.693635	0.390683
H	4.382369	-0.662280	-0.958671
H	2.067226	-1.419952	-1.383360
H	0.329251	0.827099	-1.834500
H	-1.608787	1.379440	-0.813130
H	-1.798227	1.168190	2.121325
H	-3.593649	-1.829768	2.345202
H	4.949388	-1.537797	1.318588
H	3.329975	-2.073049	0.839625
H	3.506617	-0.730940	1.993628
H	0.095888	-1.338160	-3.036623
H	-1.417513	-0.869359	-2.225146
H	-0.400965	-2.137028	-1.520022
H	-2.982154	2.952103	0.885799
H	-4.149029	1.890278	1.700926
H	-3.892354	1.729224	-0.053084
O	3.885477	2.319618	-2.055652
H	4.426862	2.651901	-2.786574
H	3.102932	2.903135	-2.008072
O	3.881603	2.078035	2.744154
H	4.252272	1.672567	3.541368
H	2.904517	2.055853	2.850900
O	1.103995	1.943622	2.606690
H	0.699607	1.208586	3.134811
H	1.264120	1.569911	1.717219
O	1.251546	3.291461	-1.606776
H	1.363969	2.636248	-0.887854
H	0.587515	3.933143	-1.267347
O	-0.122392	-0.141440	3.840430
H	-0.983688	0.176002	4.189767
H	-0.316623	-0.549217	2.975764
O	-2.623493	0.866775	4.523824
H	-2.940656	0.831456	5.438458
H	-3.174329	0.242797	4.018590

O	0.608882	-3.278689	0.696665
H	0.515218	-2.320786	0.882732
H	-0.308476	-3.578870	0.514519
O	2.354337	-3.281935	-1.323854
H	1.663000	-3.430876	-0.622050
H	2.056980	-3.754302	-2.116409
O	6.793605	-2.352399	-1.138981
H	6.069173	-2.975659	-0.889366
H	6.668311	-2.148287	-2.076623
O	7.294676	-0.165779	0.386324
H	7.643325	-0.424920	1.251397
H	7.179247	-0.998599	-0.132224
O	4.822309	-4.066475	-0.373891
H	3.938585	-3.857372	-0.753334
H	5.012907	-4.982151	-0.624783
O	-2.206244	1.705721	-2.522197
H	-1.426783	1.806530	-3.116457
H	-2.577778	2.613130	-2.409415
O	-3.887229	-0.507067	-2.904130
H	-4.747811	-0.343935	-2.460452
H	-3.329780	0.283547	-2.741258
O	0.206572	2.016562	-3.812824
H	0.261295	2.502660	-4.648566
H	0.649584	2.573569	-3.131854
O	-6.362083	-0.450674	-1.614457
H	-6.553394	0.284114	-1.013939
H	-6.185005	-1.230534	-1.044706
O	-5.277401	-2.619392	-0.264368
H	-4.458680	-2.093987	-0.130928
H	-5.025884	-3.199053	-1.006537
O	-3.937489	-3.338190	2.111406
H	-4.720818	-3.347377	1.526759
H	-3.209042	-3.641098	1.519308
O	-2.055159	-3.985761	0.158250
H	-2.403232	-3.511578	-0.637214
H	-2.101350	-4.928020	-0.068694
O	-3.253375	-3.022846	-2.121689
H	-3.373272	-2.072394	-2.384428
H	-2.991008	-3.495334	-2.925239
O	-2.971254	4.271581	-1.981673
H	-3.800351	4.400332	-1.498785
H	-2.248110	4.525663	-1.365211
O	-0.758826	4.787381	-0.408196
H	-0.566693	5.731668	-0.301917
H	-0.801383	4.410131	0.510364
O	-0.917660	3.708168	2.066442
H	-0.957104	4.376172	2.766743
H	-0.161895	3.115948	2.297740

Table S63. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_PPII-PPII_B** conformation.

N	4.836494	0.792995	0.414393
C	3.990799	-0.371271	0.046159
C	2.611196	0.171020	-0.280438
O	2.259923	1.275044	0.176468
N	1.805372	-0.584140	-1.017834
C	0.448093	-0.136392	-1.274736
C	-0.275761	0.134533	0.040122

O	-0.064075	-0.553120	1.052360
N	-1.190622	1.110108	0.012978
C	-2.219735	1.124418	1.036553
C	-2.907272	-0.240294	1.042409
O	-2.979989	-0.930911	0.035309
O	-3.454492	-0.573096	2.190761
C	3.916101	-1.363428	1.204241
C	-0.329876	-1.183309	-2.068490
C	-3.255118	2.203190	0.724740
H	4.509342	1.220156	1.308751
H	4.774866	1.508434	-0.332865
H	5.823131	0.470808	0.515318
H	4.437357	-0.839151	-0.836825
H	2.081334	-1.532553	-1.295199
H	0.490221	0.791742	-1.853174
H	-1.446134	1.480286	-0.911994
H	-1.769835	1.317590	2.015621
H	-3.882724	-1.526557	2.101053
H	4.921407	-1.696567	1.477908
H	3.336321	-2.241199	0.907863
H	3.438924	-0.900292	2.074430
H	0.188981	-1.399648	-3.006892
H	-1.326515	-0.797822	-2.294903
H	-0.443141	-2.108716	-1.494857
H	-2.764716	3.179451	0.727906
H	-4.040287	2.203309	1.485192
H	-3.712319	2.023342	-0.255006
O	4.079944	2.215542	-1.922477
H	4.658043	2.557915	-2.619862
H	3.303031	2.807806	-1.895662
O	3.934268	1.875112	2.870949
H	4.237883	1.423468	3.671740
H	2.953337	1.910832	2.928976
O	1.161006	1.916055	2.625286
H	0.687369	1.215029	3.142076
H	1.316625	1.530044	1.740497
O	1.444907	3.232850	-1.563832
H	1.515393	2.569069	-0.847634
H	0.794788	3.896307	-1.239249
O	-0.259838	-0.065265	3.821977
H	-1.118042	0.310422	4.117823
H	-0.431686	-0.472211	2.952218
O	-2.739129	1.079862	4.345015
H	-3.112927	1.050163	5.238104
H	-3.290641	0.492606	3.796345
O	0.380405	-3.280522	0.758359
H	0.364293	-2.316400	0.932430
H	-0.555233	-3.505661	0.558655
O	2.173516	-3.427194	-1.212248
H	1.452966	-3.511893	-0.528722
H	1.861349	-3.883157	-2.008573
O	6.500696	-2.629710	-1.065033
H	5.796224	-3.259939	-0.777916
H	6.374036	-2.486015	-2.013476
O	7.305417	-0.489904	0.398546
H	7.710291	-0.772866	1.230979
H	7.088707	-1.312112	-0.101799
O	4.565825	-4.339750	-0.211757
H	3.692618	-4.108759	-0.603548
H	4.736990	-5.261154	-0.455505
O	-2.033265	1.787274	-2.648781
H	-1.224876	1.866208	-3.208425
H	-2.373755	2.707719	-2.533846
O	-3.728035	-0.293654	-3.171905

H	-3.849137	-0.342094	-4.132033
H	-3.152095	0.493613	-3.008573
O	0.435365	2.022431	-3.820319
H	0.546911	2.513771	-4.647301
H	0.872406	2.553218	-3.114753
O	-5.551541	-0.710578	-1.097559
H	-5.291705	-0.532960	-2.021106
H	-4.685232	-0.795775	-0.655546
O	-6.418424	-2.981364	0.120552
H	-7.305548	-2.885419	0.495985
H	-6.269591	-2.188761	-0.446264
O	-4.320291	-2.924486	1.886472
H	-5.126355	-2.950581	1.313969
H	-3.597473	-3.297909	1.328068
O	-2.314666	-3.791905	0.171970
H	-2.601833	-3.318060	-0.650009
H	-2.413948	-4.734913	-0.032948
O	-3.239392	-2.889232	-2.227112
H	-3.315403	-1.941497	-2.470508
H	-2.772344	-3.310976	-2.963315
O	-2.703917	4.365851	-2.094314
H	-3.550523	4.532530	-1.655242
H	-2.002185	4.590985	-1.442828
O	-0.540258	4.810662	-0.430839
H	-0.318384	5.750290	-0.341681
H	-0.604434	4.454885	0.495156
O	-0.751396	3.789263	2.061673
H	-0.759060	4.469149	2.751534
H	-0.026848	3.162877	2.303157

Table S64. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 22H_2O$ peptide-water complex in the **$22H_2O$_PPII-PPII_C** conformation.

N	4.763636	0.441143	0.684944
C	3.847445	-0.599478	0.154355
C	2.550713	0.096340	-0.218443
O	2.243477	1.165541	0.341307
N	1.759964	-0.502210	-1.099497
C	0.451475	0.056785	-1.388108
C	-0.358836	0.189629	-0.100238
O	-0.251440	-0.631387	0.827433
N	-1.229505	1.199195	-0.063671
C	-2.277326	1.198290	0.940168
C	-3.009148	-0.142226	0.876248
O	-3.207706	-0.727284	-0.176674
O	-3.419660	-0.584412	2.047554
C	3.584369	-1.666055	1.215833
C	-0.301173	-0.831354	-2.375624
C	-3.255499	2.338974	0.673245
H	4.399910	0.832517	1.581613
H	4.835966	1.212504	-0.003050
H	5.703972	0.016402	0.844264
H	4.316535	-1.035732	-0.733316
H	2.011375	-1.421322	-1.480725
H	0.581644	1.051614	-1.825746
H	-1.367904	1.750513	-0.921309
H	-1.834034	1.316096	1.933968
H	-3.769638	-1.561371	1.950003
H	4.524752	-2.134057	1.520408
H	2.930067	-2.442456	0.810929

H	3.101606	-1.219057	2.091583
H	0.278929	-0.935409	-3.297183
H	-1.265052	-0.377196	-2.617426
H	-0.482655	-1.823872	-1.950195
H	-2.719242	3.289223	0.733910
H	-4.049597	2.339523	1.424587
H	-3.707798	2.232232	-0.318567
O	4.300708	2.085710	-1.570604
H	4.954148	2.381651	-2.221309
H	3.620275	2.785698	-1.527229
O	3.777072	1.439812	3.146354
H	4.031165	0.958495	3.947085
H	2.797074	1.517452	3.167274
O	1.031120	1.639029	2.758481
H	0.506373	0.895279	3.153025
H	1.239341	1.355939	1.845681
O	1.804887	3.378493	-1.220832
H	1.759414	2.630495	-0.590501
H	1.202459	4.063433	-0.851990
O	-0.516207	-0.401075	3.640515
H	-1.340537	0.021857	3.966315
H	-0.708680	-0.695859	2.730618
O	-2.847005	0.981257	4.286530
H	-3.241442	0.940505	5.170228
H	-3.431206	0.468193	3.699996
O	0.194866	-3.301271	0.221793
H	0.177432	-2.362828	0.506103
H	-0.727890	-3.476865	-0.063963
O	2.139166	-3.295442	-1.600776
H	1.358901	-3.439865	-0.997822
H	1.897233	-3.657720	-2.466754
O	6.611853	-2.938506	-0.983323
H	5.799629	-3.484232	-0.848692
H	6.569992	-2.603805	-1.890454
O	7.128015	-1.010395	0.852483
H	7.362420	-1.413319	1.700849
H	6.993951	-1.754140	0.215698
O	4.394748	-4.440680	-0.504721
H	3.577575	-4.100499	-0.934954
H	4.494357	-5.356763	-0.802104
O	-1.707774	2.438788	-2.590904
H	-0.864967	2.489044	-3.100436
H	-1.962160	3.369918	-2.381159
O	-3.570811	0.515839	-3.205405
H	-3.836453	0.671546	-4.123503
H	-2.924458	1.227583	-2.989873
O	0.823348	2.513442	-3.632563
H	1.022606	3.079985	-4.392311
H	1.267954	2.919142	-2.851982
O	-5.665777	-1.826385	-1.076668
H	-5.220987	-2.209166	-1.856208
H	-4.920141	-1.396231	-0.613635
O	-6.521720	-3.552837	0.824453
H	-7.281621	-3.196974	1.307009
H	-6.371612	-2.950698	0.055911
O	-4.009987	-3.023408	1.754544
H	-4.949476	-3.241972	1.540944
H	-3.488015	-3.256303	0.951063
O	-2.502081	-3.562859	-0.522551
H	-2.695631	-4.438667	-0.892229
H	-2.775173	-2.920823	-1.221642
O	-3.460693	-2.173272	-2.710323
H	-3.389789	-1.194420	-2.825411
H	-3.192722	-2.567308	-3.553507

O	-2.191070	4.998994	-1.791906
H	-3.040946	5.170477	-1.361144
H	-1.502098	5.088170	-1.095451
O	-0.083951	5.020052	-0.009494
H	0.230337	5.904371	0.233258
H	-0.293903	4.556964	0.844572
O	-0.712480	3.680220	2.248516
H	-0.830843	4.233216	3.034699
H	-0.058706	2.982834	2.497378

Table S65. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 8H_2O$ peptide-water complex in the **$8H_2O$_β-PPII** conformation.

N	3.494587	-0.191155	0.210119
C	2.286821	0.236563	-0.539451
C	1.140801	-0.692038	-0.156007
N	-0.056206	-0.124435	-0.066994
C	-1.256370	-0.884637	0.241055
C	-2.456014	-0.100379	-0.276393
N	-3.522907	-0.825730	-0.647457
C	-4.777613	-0.179224	-0.966528
C	-5.328051	0.526549	0.269147
O	-6.250648	1.432395	-0.073459
O	1.346100	-1.907703	0.006954
O	-2.463534	1.133533	-0.299391
C	-5.791620	-1.201819	-1.479622
O	-5.011019	0.288043	1.409917
C	2.553946	0.167925	-2.040438
C	-1.409081	-1.092754	1.753504
H	4.265785	0.483300	0.008371
H	3.803311	-1.145602	-0.082241
H	3.282744	-0.191847	1.225365
H	2.058587	1.262723	-0.233989
H	-0.134799	0.887653	-0.193890
H	-1.183964	-1.852206	-0.264988
H	-3.498763	-1.835781	-0.568082
H	-4.599361	0.585027	-1.729426
H	-6.614663	1.835627	0.736490
H	-5.990463	-1.965029	-0.720795
H	-5.402483	-1.685935	-2.378595
H	-6.729929	-0.703580	-1.730371
H	1.679561	0.522247	-2.591835
H	2.770035	-0.862114	-2.342270
H	3.410193	0.798700	-2.293938
H	-2.306153	-1.677249	1.975403
H	-1.481901	-0.124322	2.257673
H	-0.537223	-1.629846	2.136344
O	2.299701	-0.238001	2.757101
H	2.022922	-1.171001	2.895885
H	1.529154	0.315915	2.948955
O	1.826508	-2.922871	2.556580
H	1.563752	-2.849704	1.619082
H	1.177297	-3.498342	2.985955
O	4.465029	-2.670812	-0.714547
H	5.187471	-2.572013	-1.351647
H	3.764095	-3.193617	-1.166808
O	2.224634	-3.911506	-1.703150
H	2.020427	-3.836929	-2.646693
H	1.667442	-3.250181	-1.249388
O	-0.234485	2.772018	-0.534078

H	-0.075452	3.085496	-1.437569
H	-1.186750	2.558353	-0.479459
O	5.240925	1.792452	-0.654809
H	6.077264	1.950888	-0.193498
H	4.743635	2.646654	-0.627963
O	3.733660	4.049068	-0.578667
H	3.342123	4.284763	-1.432018
H	2.985243	3.988513	0.060153
O	1.640575	3.856767	1.190865
H	0.866888	3.490842	0.713598
H	1.799191	3.269656	1.944356

Table S66. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 11H_2O$ peptide-water complex in the **$11H_2O$_β-PPII** conformation.

N	3.778303	-0.448509	0.465999
C	2.600359	-0.070190	-0.360609
C	1.401521	-0.860577	0.152704
N	0.283059	-0.167477	0.349804
C	-0.961697	-0.761570	0.798217
C	-2.013460	-0.664314	-0.301509
N	-2.971480	-1.599673	-0.293753
C	-4.179710	-1.422209	-1.072524
C	-4.934245	-0.199536	-0.556643
O	-5.912738	0.154161	-1.383937
O	1.511742	-2.084318	0.345131
O	-2.030879	0.277421	-1.108444
C	-5.058918	-2.670146	-0.999170
O	-4.688763	0.369375	0.484932
C	2.868192	-0.398774	-1.825385
C	-1.485323	-0.013849	2.030794
H	4.595007	0.128645	0.166191
H	4.030405	-1.452374	0.327517
H	3.564544	-0.273518	1.464638
H	2.438012	1.004037	-0.223152
H	0.311845	0.857217	0.259149
H	-0.769318	-1.808591	1.042255
H	-2.960459	-2.317949	0.422655
H	-3.905141	-1.219290	-2.112411
H	-6.387443	0.922856	-1.014804
H	-5.360791	-2.874832	0.032628
H	-4.504909	-3.529175	-1.384830
H	-5.955570	-2.530599	-1.605413
H	2.015239	-0.081227	-2.429731
H	3.026872	-1.474901	-1.948974
H	3.763018	0.129692	-2.166674
H	-2.452337	-0.416351	2.344560
H	-1.610818	1.050269	1.801913
H	-0.769421	-0.127691	2.847838
O	2.462842	0.064623	2.898328
H	1.850696	-0.683183	3.085242
H	1.932733	0.873416	2.951842
O	0.869916	-2.176947	3.046824
H	1.128889	-2.529194	2.173459
H	1.142718	-2.837632	3.700682
O	4.692495	-3.058107	-0.106498
H	5.435908	-3.015640	-0.725467
H	3.996692	-3.598751	-0.545450
O	2.437144	-4.280468	-1.064036
H	2.259448	-4.312796	-2.015173

H	1.907051	-3.540267	-0.708416
O	0.332588	2.675376	-0.070524
H	0.373422	2.505663	-1.038739
H	-0.608468	2.929891	0.092409
O	5.688477	1.164646	-0.751939
H	6.450394	1.518583	-0.270676
H	5.177912	1.944083	-1.081143
O	4.063734	3.180892	-1.561247
H	3.423167	2.983189	-2.260070
H	3.542269	3.473347	-0.775198
O	2.650004	3.877108	0.668763
H	1.748497	3.508773	0.517514
H	2.997161	3.431699	1.455401
O	-2.368488	2.929397	-0.054506
H	-2.945925	2.931430	0.740075
H	-2.477101	2.045186	-0.452160
O	0.155642	1.413689	-2.474797
H	-0.635249	0.942013	-2.141436
H	-0.093834	1.788529	-3.332413
O	-4.160664	2.652087	2.048623
H	-4.548532	1.827919	1.703001
H	-3.800475	2.432637	2.920377

Table S67. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 5H_2O$ peptide-water complex in the **$5H_2O$_PPII-β** conformation.

N	-4.061590	0.346740	-0.174830
C	-3.079390	0.014880	-1.241240
C	-1.847380	-0.561050	-0.559560
N	-1.108900	-1.410110	-1.275650
C	0.215880	-1.809650	-0.838850
C	1.103910	-0.562880	-0.693970
N	2.225760	-0.738810	0.015840
C	3.222710	0.306520	0.153970
C	4.563300	-0.363750	0.387840
O	5.562330	0.517660	0.356580
O	-1.539120	-0.197500	0.584590
O	0.802340	0.506280	-1.231240
C	2.901000	1.273100	1.300890
O	4.708570	-1.545090	0.607120
C	-2.693210	1.271060	-2.020790
C	0.815260	-2.782740	-1.854410
H	-4.950650	0.640590	-0.588960
H	-3.715770	1.138580	0.424560
H	-4.236590	-0.494010	0.424640
H	-3.541500	-0.728180	-1.895780
H	-1.386470	-1.632000	-2.225860
H	0.131550	-2.294040	0.140240
H	2.443170	-1.642660	0.425820
H	3.277950	0.859630	-0.790280
H	6.401370	0.056700	0.544010
H	2.910770	0.742800	2.257910
H	1.908260	1.702320	1.147160
H	3.633560	2.083130	1.330990
H	-2.006430	1.010620	-2.829120
H	-2.200630	1.993640	-1.362030
H	-3.583320	1.731490	-2.456990
H	1.798380	-3.123380	-1.523910
H	0.925010	-2.299460	-2.830180

H	0.166310	-3.656030	-1.958300
O	-3.205380	2.555560	1.269900
H	-3.590770	3.384610	0.950540
H	-2.223130	2.653890	1.203790
O	-0.516810	2.386570	1.077690
H	0.002070	2.832170	0.373040
H	-0.568600	1.450080	0.810030
O	1.197710	3.214830	-0.944950
H	0.852260	3.734750	-1.685120
H	1.210910	2.284510	-1.249150
O	-4.243850	-2.020260	1.262170
H	-3.458410	-2.125680	1.847870
H	-4.307000	-2.828970	0.733590
O	-1.964180	-1.874150	2.769640
H	-1.526050	-1.287260	2.124220
H	-1.378530	-2.636820	2.884410

Table S68. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 13H_2O$ peptide-water complex in the 13**H_2O_PPII-β** conformation.

N	-3.255587	0.320657	-1.609277
C	-2.694974	-0.083551	-0.296709
C	-1.232946	0.339672	-0.252472
O	-0.543799	0.349816	-1.285768
N	-0.740013	0.630338	0.950502
C	0.684966	0.807911	1.155974
C	1.438807	-0.452731	0.714782
O	0.883579	-1.566838	0.711844
N	2.719318	-0.277240	0.396787
C	3.569455	-1.398970	0.042142
C	5.001372	-1.055972	0.394302
O	5.408309	0.069883	0.593662
O	5.775608	-2.135418	0.409103
C	-2.799697	-1.596692	-0.119292
C	0.953810	1.085021	2.635135
C	3.482965	-1.731664	-1.454679
H	-2.831301	-0.228364	-2.388339
H	-3.085088	1.337033	-1.754247
H	-4.287006	0.151206	-1.615571
H	-3.265273	0.436068	0.479970
H	-1.353095	0.560910	1.768927
H	1.037161	1.651106	0.551915
H	3.122125	0.666399	0.442729
H	3.265235	-2.267582	0.634213
H	6.697737	-1.868149	0.585216
H	-3.841431	-1.911080	-0.227219
H	-2.456401	-1.884699	0.877796
H	-2.186430	-2.112127	-0.866445
H	0.406660	1.977574	2.950901
H	2.020002	1.252014	2.800513
H	0.635071	0.236676	3.250478
H	2.442965	-1.936461	-1.720621
H	4.088615	-2.611114	-1.686925
H	3.833548	-0.883127	-2.050326
O	-2.587192	3.033459	-1.468212
H	-2.791845	3.425139	-0.606824
H	-1.613646	3.149403	-1.605234
O	-2.330424	-1.393130	-3.655271
H	-2.930524	-2.147887	-3.742885

H	-1.452754	-1.762676	-3.391508
O	0.134090	-1.985440	-2.705566
H	0.260634	-2.753281	-2.106705
H	0.087907	-1.203334	-2.122808
O	0.088002	3.043866	-1.905042
H	0.179050	2.080463	-1.786735
H	0.562988	3.461178	-1.150649
O	0.663787	-3.895032	-0.755407
H	1.448373	-4.445401	-0.895501
H	0.921482	-3.207792	-0.109246
O	-0.903341	-2.314401	2.706515
H	-0.273706	-2.057341	1.997420
H	-0.368467	-2.707716	3.411890
O	-2.444401	-0.155867	3.122836
H	-1.900181	-0.980572	3.183865
H	-2.389306	0.286544	3.983622
O	-6.517113	1.013599	1.043429
H	-5.936917	0.433437	1.590729
H	-6.190085	1.917475	1.157154
O	-6.019225	-0.016916	-1.423369
H	-6.372181	-0.918087	-1.439237
H	-6.277803	0.367274	-0.550050
O	-4.968614	-0.849075	2.269166
H	-4.102924	-0.609445	2.668925
H	-5.437235	-1.374435	2.933883
O	5.464470	2.328657	-1.095199
H	5.850388	1.538579	-0.678433
H	6.178732	2.979476	-1.159766
O	3.476981	2.540894	0.740251
H	4.192344	2.617579	0.061553
H	3.929850	2.619773	1.593998
O	1.379525	4.235437	0.214687
H	2.164750	3.715983	0.497908
H	0.847481	4.387009	1.009199

Table S69. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 17H_2O$ peptide-water complex in the 17**H_2O_PPII-β** conformation.

N	4.565795	-0.003516	-1.339400
C	3.795567	0.045981	-0.072734
C	2.344900	-0.281794	-0.391543
O	1.855483	0.034476	-1.491469
N	1.635196	-0.848262	0.580171
C	0.193306	-0.970762	0.475570
C	-0.439244	0.415531	0.296268
O	0.132628	1.441709	0.712125
N	-1.639388	0.422085	-0.271681
C	-2.444941	1.627281	-0.367927
C	-3.907224	1.227544	-0.269794
O	-4.278833	0.085612	-0.505876
O	-4.690290	2.224971	0.047017
C	3.883463	1.439310	0.547214
C	-0.359766	-1.635112	1.736097
C	-2.199063	2.384711	-1.678582
H	4.274906	0.763008	-1.986337
H	4.410685	-0.923082	-1.792304
H	5.586403	0.104430	-1.137181
H	4.217113	-0.704845	0.602993
H	2.088782	-1.047929	1.478027
H	-0.054466	-1.578369	-0.401250

H	-2.063063	-0.473691	-0.545268
H	-2.206658	2.270683	0.484260
H	-5.633577	1.888572	0.288380
H	4.928789	1.694100	0.741071
H	3.344983	1.462296	1.498268
H	3.443868	2.183998	-0.125175
H	0.106181	-2.614551	1.875442
H	-1.439498	-1.773163	1.642828
H	-0.158561	-1.017223	2.618186
H	-1.139566	2.640809	-1.763315
H	-2.789891	3.304149	-1.700500
H	-2.474966	1.760016	-2.533557
O	3.742305	-2.613223	-2.002059
H	3.774550	-3.277743	-1.298815
H	2.834622	-2.643611	-2.370292
O	3.952238	2.243007	-2.929726
H	4.561537	2.969090	-2.732190
H	3.046624	2.568079	-2.703584
O	1.371618	2.704626	-2.238930
H	1.154982	3.300713	-1.489220
H	1.301779	1.799316	-1.881535
O	1.128164	-2.226256	-2.879217
H	1.159966	-1.301045	-2.556404
H	0.940698	-2.187370	-3.829583
O	0.556512	4.077095	0.034634
H	-0.197969	4.675956	-0.065957
H	0.202680	3.247387	0.413501
O	1.523043	1.472319	3.109651
H	1.027792	1.464727	2.260648
H	0.876499	1.690878	3.797077
O	2.930184	-0.810318	3.138024
H	2.394270	-0.001879	3.338649
H	2.717558	-1.468498	3.817303
O	7.360681	-1.460739	1.544451
H	6.679391	-1.045323	2.124450
H	7.059473	-2.363889	1.369932
O	7.261693	0.130295	-0.654882
H	7.621940	0.995795	-0.413504
H	7.374886	-0.449581	0.138181
O	5.566015	-0.015664	2.984777
H	4.645878	-0.331749	3.126295
H	5.894921	0.252213	3.855207
O	-6.403936	-1.635110	-0.844730
H	-5.751400	-0.900958	-0.832262
H	-6.329252	-2.048387	-1.718500
O	-2.550958	-2.285576	-0.782866
H	-3.105798	-2.208758	-1.574806
H	-1.862908	-2.962932	-0.993236
O	-0.489098	-3.942496	-1.406884
H	0.028205	-4.338097	-0.690263
H	0.136863	-3.410172	-1.942808
O	-4.594107	-2.528646	1.144338
H	-3.803531	-2.450837	0.569639
H	-5.343008	-2.488930	0.517913
O	-8.660060	-0.153063	-0.341199
H	-8.007280	-0.847631	-0.578117
H	-9.330112	-0.582646	0.209899
O	-6.846647	1.361682	1.061134
H	-7.546678	0.872610	0.569848
H	-6.372231	0.711241	1.632249
O	-5.036075	-0.144400	2.419269
H	-5.186017	-0.371636	3.348483
H	-4.818077	-0.996221	1.965521

Table S70. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 19H_2O$ peptide-water complex in the **$19H_2O$_PPII-β** conformation.

N	4.740231	0.039107	-1.194376
C	3.885455	-0.004420	0.017375
C	2.490354	-0.441038	-0.402182
O	2.045301	-0.139444	-1.524402
N	1.772571	-1.088092	0.511685
C	0.351223	-1.312432	0.324468
C	-0.369432	0.029488	0.145899
O	0.082085	1.071152	0.656392
N	-1.515135	-0.021346	-0.526457
C	-2.408986	1.119996	-0.603758
C	-3.837020	0.609487	-0.564465
O	-4.130961	-0.543778	-0.827843
O	-4.711715	1.548791	-0.260641
C	3.816757	1.376959	0.665915
C	-0.214317	-2.041555	1.542737
C	-2.178783	1.961768	-1.864252
H	4.439128	0.801976	-1.841380
H	4.685922	-0.876262	-1.677541
H	5.733589	0.216689	-0.919115
H	4.321805	-0.734529	0.706352
H	2.187926	-1.265179	1.432259
H	0.193909	-1.915045	-0.576612
H	-1.843357	-0.930124	-0.875753
H	-2.248199	1.734555	0.288502
H	-5.563579	1.132266	0.156530
H	4.824913	1.722019	0.910679
H	3.236593	1.331401	1.591554
H	3.341696	2.093840	-0.012811
H	0.293531	-3.001325	1.672212
H	-1.282437	-2.227234	1.410421
H	-0.075721	-1.440560	2.448440
H	-1.145920	2.319247	-1.881809
H	-2.847245	2.827096	-1.875985
H	-2.357507	1.358277	-2.759544
O	4.156618	-2.609472	-1.958656
H	4.197267	-3.287196	-1.268551
H	3.280227	-2.702735	-2.386909
O	4.087203	2.284381	-2.764656
H	4.600986	3.050494	-2.470201
H	3.135014	2.514308	-2.621371
O	1.431617	2.492446	-2.320182
H	1.113533	3.037019	-1.560530
H	1.406466	1.570440	-2.002618
O	1.577536	-2.409449	-2.999743
H	1.514155	-1.495704	-2.649424
H	1.450090	-2.352307	-3.959149
O	0.419445	3.739527	-0.108996
H	-0.401422	4.244080	-0.311916
H	0.133480	2.897592	0.293728
O	1.309958	1.135920	3.128796
H	0.862615	1.118521	2.253167
H	0.607724	1.235991	3.788466
O	2.940296	-0.991738	3.129960
H	2.318719	-0.247140	3.333278
H	2.759097	-1.696037	3.771152
O	7.486693	-1.205389	1.880747
H	6.742307	-0.852329	2.423719
H	7.273533	-2.130656	1.692547

O	7.366957	0.372066	-0.327328
H	7.643598	1.263192	-0.069504
H	7.483170	-0.197564	0.472789
O	5.491127	0.054697	3.226840
H	4.595400	-0.348180	3.270640
H	5.718224	0.305006	4.134223
O	-6.019307	-2.533138	-0.489687
H	-5.501515	-1.744658	-0.758503
H	-6.070778	-3.105861	-1.269907
O	-2.185021	-2.766393	-1.137868
H	-2.824891	-2.784407	-1.866082
H	-1.464797	-3.403251	-1.369993
O	-0.021407	-4.285760	-1.725343
H	0.459676	-4.664051	-0.974833
H	0.605800	-3.688231	-2.186674
O	-3.751877	-2.895380	1.167391
H	-3.157391	-2.893318	0.385991
H	-4.642009	-2.984716	0.773790
O	-8.263613	-1.363251	0.556374
H	-7.596988	-1.965060	0.158633
H	-8.645716	-1.837770	1.308865
O	-6.447574	0.558737	1.244140
H	-7.144164	-0.106097	1.026679
H	-5.734431	0.102670	1.745831
O	-3.965208	-0.308911	2.055399
H	-3.665612	-0.268436	2.975282
H	-3.816521	-1.241700	1.760501
O	-1.873636	5.146680	-0.718723
H	-1.794899	6.084533	-0.491196
H	-2.619036	4.798969	-0.177482
O	-3.902383	4.052394	0.787887
H	-3.639330	3.891698	1.706090
H	-4.248869	3.201057	0.457853

Table S71. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 5H_2O$ peptide-water complex in the **$5H_2O$_PPII-PPII** conformation.

N	0.167414	-0.218337	0.092750
C	0.110265	1.267931	0.077807
C	1.543741	1.767709	-0.000524
O	2.464293	1.090389	0.480346
N	1.745851	2.975907	-0.527995
C	3.034372	3.632710	-0.371519
C	3.383798	3.670231	1.119687
O	2.510284	3.874654	1.965820
N	4.676558	3.498767	1.434761
C	5.146068	3.670228	2.793257
C	4.974653	5.122887	3.228094
O	4.770899	6.056144	2.489280
O	5.109508	5.238059	4.553751
C	-0.538396	1.785868	1.361082
C	2.964900	5.057321	-0.915783
C	6.610214	3.246465	2.909760
H	0.641129	-0.565750	0.964580
H	0.692629	-0.560323	-0.746047
H	-0.776387	-0.613816	0.070232
H	-0.458309	1.569957	-0.805047
H	0.959887	3.518647	-0.869853
H	3.793546	3.057180	-0.912443
H	5.359093	3.376969	0.695118

H	4.536025	3.052488	3.461781
H	5.031934	6.178433	4.800659
H	-1.554870	1.394799	1.454757
H	-0.586627	2.876743	1.342198
H	0.047570	1.476759	2.233102
H	2.696464	5.041785	-1.975277
H	3.937168	5.544318	-0.811283
H	2.221522	5.639627	-0.362830
H	6.711158	2.197965	2.618744
H	6.948473	3.356816	3.941433
H	7.245981	3.859533	2.263474
O	1.692828	-0.749997	-2.163370
H	1.492558	-0.191182	-2.928249
H	2.645625	-0.609621	-1.954644
O	1.301846	-1.114860	2.469164
H	0.665072	-1.324446	3.167932
H	1.948245	-0.475081	2.856390
O	3.133128	0.777373	3.195447
H	2.749100	1.476842	3.769586
H	3.092076	1.135025	2.289735
O	4.211862	-0.312073	-1.175427
H	3.884467	0.299158	-0.488849
H	4.866769	0.183039	-1.689203
O	2.178210	2.994205	4.557836
H	1.263363	2.963489	4.873485
H	2.165756	3.508311	3.725436

Table S72. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 6H_2O$ peptide-water complex in the **$6H_2O$_PPII-PPII** conformation.

N	-3.966668	1.088868	-0.489315
C	-3.071367	0.359047	-1.425439
C	-2.083932	-0.423595	-0.575594
O	-1.749792	0.003516	0.538820
N	-1.545708	-1.521983	-1.108967
C	-0.391049	-2.140620	-0.477428
C	0.723183	-1.095417	-0.365626
O	0.939317	-0.302761	-1.283746
N	1.457453	-1.118803	0.760760
C	2.664813	-0.329366	0.869603
C	3.701853	-0.837768	-0.128845
O	3.669928	-1.900361	-0.695728
O	4.700585	0.050097	-0.282628
C	-2.308145	1.345158	-2.309191
C	0.098520	-3.316826	-1.318080
C	3.220684	-0.380059	2.292378
H	-3.426733	1.798542	0.069247
H	-4.418592	0.406332	0.161450
H	-4.699101	1.583541	-1.005092
H	-3.691764	-0.311843	-2.024088
H	-1.804232	-1.813316	-2.045753
H	-0.678115	-2.483199	0.522227
H	1.277842	-1.829650	1.461219
H	2.431725	0.710546	0.615109
H	5.368768	-0.318233	-0.891300
H	-3.008206	1.956541	-2.884538
H	-1.668835	0.800898	-3.007766
H	-1.679323	1.998122	-1.695398
H	-0.692153	-4.065644	-1.412501
H	0.963651	-3.782007	-0.840050

H	0.393489	-2.974753	-2.314713
H	2.470792	0.003969	2.988140
H	4.115473	0.241212	2.365464
H	3.479881	-1.405208	2.574272
O	-4.835925	-1.029431	1.081431
H	-5.017736	-1.854263	0.608227
H	-4.157066	-1.239371	1.763834
O	-2.530039	3.027919	0.887425
H	-2.554526	3.911023	0.491040
H	-1.574349	2.788495	0.983173
O	0.027998	2.100924	1.122874
H	0.530454	2.308775	0.298960
H	-0.287818	1.187379	1.005022
O	-2.742058	-1.229277	2.838216
H	-2.118329	-0.842467	2.195054
H	-2.373934	-2.089400	3.088556
O	1.491770	2.454854	-1.171773
H	2.397495	2.615576	-0.830339
H	1.459178	1.511876	-1.422408
O	3.912345	2.750167	0.170146
H	4.437106	3.555798	0.053534
H	4.509037	2.005655	-0.018879

Table S73. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 8H_2O$ peptide-water complex in the **$8H_2O$_PPII-PPII** conformation.

N	3.820916	-1.067921	-0.843388
C	3.052601	0.200491	-0.960362
C	2.010011	0.193074	0.145946
O	1.561963	-0.884381	0.573735
N	1.562169	1.377193	0.551995
C	0.389570	1.455864	1.403412
C	-0.778366	0.755291	0.706531
O	-0.983898	0.909276	-0.506697
N	-1.580693	0.006199	1.470676
C	-2.821866	-0.528050	0.950481
C	-3.780958	0.619923	0.647119
O	-3.648994	1.760199	1.012053
O	-4.832205	0.190528	-0.073033
C	2.351378	0.275851	-2.315325
C	0.024664	2.918432	1.650542
C	-3.445355	-1.512641	1.938987
H	3.199442	-1.897045	-1.017121
H	4.236613	-1.146117	0.113630
H	4.577445	-1.093550	-1.531903
H	3.747065	1.034342	-0.832597
H	1.946193	2.232025	0.125165
H	0.598523	0.952056	2.353301
H	-1.399165	-0.063665	2.466095
H	-2.612051	-1.050202	0.010143
H	-5.443166	0.935756	-0.228095
H	3.081961	0.192364	-3.124764
H	1.837773	1.235209	-2.412700
H	1.616393	-0.530723	-2.411641
H	0.851178	3.427106	2.153171
H	-0.863345	2.981914	2.283672
H	-0.182828	3.425963	0.702863
H	-2.742550	-2.327428	2.129114
H	-4.361428	-1.932374	1.519220
H	-3.686400	-1.018110	2.884960

O	4.677907	-1.020455	1.803975
H	4.984138	-0.155935	2.114257
H	3.930876	-1.281083	2.391157
O	2.187977	-3.244875	-1.443972
H	2.215568	-3.526361	-2.370035
H	1.237474	-3.076571	-1.228313
O	-0.331435	-2.562381	-0.631138
H	-0.792456	-2.062489	-1.345688
H	0.063578	-1.883321	-0.054725
O	2.381804	-1.842319	3.058551
H	1.826874	-1.537411	2.315243
H	2.055307	-1.382731	3.846016
O	-1.683486	-1.030535	-2.470002
H	-2.614523	-1.320641	-2.358548
H	-1.598658	-0.199347	-1.968318
O	-4.187829	-2.013143	-1.768842
H	-4.747215	-2.433887	-2.438335
H	-4.733397	-1.324420	-1.351686
O	0.064993	2.886540	-2.174896
H	-0.309868	2.205032	-1.576238
H	-0.623914	3.559520	-2.277775
O	2.509892	3.446151	-1.101837
H	1.646973	3.484725	-1.573058
H	2.697260	4.342842	-0.788613

Table S74. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 11H_2O$ peptide-water complex in the **$11H_2O$_PPII-PPII** conformation.

N	2.990008	-1.645990	-0.251527
C	2.434835	-0.273823	-0.162149
C	1.167628	-0.341570	0.675245
O	0.484365	-1.378814	0.695372
N	0.798043	0.772331	1.303540
C	-0.510877	0.855203	1.926445
C	-1.581349	0.535796	0.881627
O	-1.499444	0.982411	-0.272504
N	-2.619737	-0.204992	1.281220
C	-3.781266	-0.383123	0.434183
C	-4.485069	0.959788	0.254196
O	-4.273085	1.954473	0.900020
O	-5.411542	0.893413	-0.717579
C	2.096055	0.249038	-1.557415
C	-0.740154	2.265324	2.467268
C	-4.733269	-1.419358	1.029822
H	2.333744	-2.277224	-0.764009
H	3.144452	-2.015680	0.707523
H	3.902711	-1.621916	-0.759736
H	3.182852	0.361911	0.322331
H	1.372429	1.617686	1.217053
H	-0.568896	0.125128	2.741196
H	-2.665214	-0.520802	2.243961
H	-3.450337	-0.728384	-0.552072
H	-5.866616	1.754018	-0.788024
H	2.990389	0.239171	-2.186358
H	1.727079	1.276629	-1.500549
H	1.322688	-0.376212	-2.017200
H	0.026312	2.506733	3.208421
H	-1.721067	2.329744	2.943531
H	-0.696496	2.999459	1.656144
H	-4.209917	-2.372216	1.139582

H	-5.586327	-1.564882	0.364560
H	-5.101061	-1.096197	2.008352
O	3.144332	-2.186427	2.510007
H	3.392319	-1.432485	3.064336
H	2.263920	-2.489501	2.828335
O	1.276344	-3.272116	-1.798337
H	1.493580	-3.266517	-2.741775
H	0.335164	-2.979646	-1.726452
O	-1.244813	-2.338722	-1.286128
H	-1.499476	-1.609178	-1.898440
H	-0.890703	-1.898243	-0.492145
O	0.560991	-3.027712	2.941382
H	0.234521	-2.483386	2.199384
H	0.066978	-2.742672	3.724065
O	-2.071770	-0.190498	-2.797514
H	-3.028725	-0.381203	-2.900872
H	-2.001100	0.436374	-2.054633
O	-4.729669	-1.006066	-2.722608
H	-5.188867	-1.253773	-3.538770
H	-5.290987	-0.345197	-2.282681
O	0.095384	3.079359	-1.144400
H	-0.476632	2.345926	-0.827657
H	-0.485108	3.846954	-1.255071
O	2.227842	3.114646	0.483544
H	1.457669	3.316125	-0.105779
H	2.318363	3.860969	1.095678
O	6.315941	0.974549	-0.336634
H	5.636976	1.655137	-0.559592
H	6.336095	0.914323	0.629232
O	5.469616	-1.335177	-1.488151
H	5.492708	-1.231283	-2.450249
H	5.840766	-0.501599	-1.107382
O	4.411260	2.730456	-1.167166
H	3.662713	2.932208	-0.562669
H	4.769677	3.586409	-1.443971

Table S75. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 13H_2O$ peptide-water complex in the 13**H_2O_PPII-PPII** conformation.

N	3.239647	0.282621	-1.722282
C	2.736292	0.180951	-0.330070
C	1.342560	-0.418821	-0.405765
O	0.638288	-0.231221	-1.411588
N	0.908891	-1.074746	0.666991
C	-0.451199	-1.584084	0.726862
C	-1.431112	-0.418962	0.587111
O	-1.356656	0.568334	1.339393
N	-2.385907	-0.531591	-0.338517
C	-3.451398	0.444604	-0.427734
C	-4.306292	0.368106	0.830718
O	-4.441788	-0.619705	1.518802
O	-4.946708	1.513893	1.073894
C	2.675733	1.563244	0.315741
C	-0.673282	-2.283254	2.065700
C	-4.318919	0.171773	-1.656933
H	2.661263	0.957915	-2.270488
H	3.180731	-0.653339	-2.158931
H	4.233766	0.604607	-1.716521
H	3.407840	-0.483951	0.221630

H	1.528775	-1.194713	1.474680
H	-0.607736	-2.290246	-0.097562
H	-2.409824	-1.352946	-0.951308
H	-3.026565	1.451097	-0.500588
H	-5.530376	1.404977	1.849609
H	3.673524	2.009754	0.340184
H	2.306746	1.482741	1.342140
H	2.000479	2.214524	-0.249786
H	0.041608	-3.104035	2.172186
H	-1.684379	-2.693817	2.108399
H	-0.533082	-1.579378	2.892501
H	-3.698522	0.230471	-2.554879
H	-5.111973	0.918962	-1.730850
H	-4.772923	-0.822571	-1.598827
O	2.640191	-2.458215	-1.863698
H	3.248477	-3.182291	-2.070451
H	1.759229	-2.725401	-2.209233
O	1.714244	2.244539	-3.072132
H	2.065712	3.144720	-3.014478
H	0.786689	2.284407	-2.734972
O	-0.843834	2.030119	-2.082513
H	-0.956091	2.541222	-1.247067
H	-0.613845	1.127642	-1.794695
O	0.045570	-2.586078	-2.796832
H	0.001109	-1.662602	-2.485092
H	-0.750600	-3.010656	-2.424214
O	-1.311719	3.244883	0.348683
H	-2.231862	3.562926	0.226598
H	-1.382569	2.427653	0.876271
O	-3.948678	3.794371	-0.336273
H	-4.290713	4.699715	-0.375996
H	-4.564564	3.295340	0.226160
O	0.631405	1.089135	3.201612
H	-0.075538	0.871753	2.553715
H	0.181253	1.293699	4.034447
O	2.537950	-0.783671	3.001227
H	1.858216	-0.121365	3.285712
H	2.560389	-1.476448	3.678800
O	6.662516	-0.342121	0.759537
H	6.027056	-0.091860	1.471988
H	6.578192	-1.299330	0.642606
O	5.912935	1.019531	-1.465708
H	6.106870	1.959194	-1.337574
H	6.246519	0.552361	-0.660216
O	4.911138	0.574251	2.626871
H	4.093763	0.062288	2.818785
H	5.319811	0.757857	3.485317
O	-2.519312	-3.077177	-1.705129
H	-3.152420	-3.162253	-2.434018
H	-2.991247	-3.367336	-0.889071
O	-3.614880	-3.254898	0.770785
H	-3.950629	-2.372024	1.027083
H	-4.290581	-3.892360	1.045447

Table S76. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 19H_2O$ peptide-water complex in the **$19H_2O$_PPII-PPII_A** conformation.

N	-4.717863	-0.753834	0.556595
C	-3.695702	0.148842	-0.026971
C	-2.414511	-0.649927	-0.192006

O	-2.186539	-1.630145	0.540010
N	-1.543902	-0.209426	-1.095361
C	-0.199820	-0.754190	-1.128014
C	0.463672	-0.567090	0.234890
O	0.193765	0.413195	0.951753
N	1.394162	-1.461617	0.571143
C	2.333736	-1.121063	1.621110
C	3.046342	0.172729	1.230224
O	3.231309	0.486676	0.064109
O	3.474425	0.881491	2.256112
C	-3.452014	1.339768	0.898201
C	0.634826	-0.034760	-2.186640
C	3.363400	-2.236279	1.797733
H	-4.448378	-1.047708	1.522840
H	-4.803453	-1.589668	-0.047331
H	-5.635746	-0.256470	0.595058
H	-4.062675	0.479534	-1.003553
H	-1.746709	0.649320	-1.618584
H	-0.244898	-1.822215	-1.362457
H	1.684526	-2.152431	-0.140142
H	1.797000	-0.959921	2.561653
H	3.907328	1.752059	1.921559
H	-4.382452	1.895090	1.046734
H	-2.718185	2.016908	0.453151
H	-3.073376	0.998545	1.867492
H	0.167165	-0.150930	-3.168466
H	1.640343	-0.460845	-2.218674
H	0.721544	1.032095	-1.952385
H	2.854833	-3.162971	2.074785
H	4.070379	-1.974623	2.588494
H	3.914672	-2.399009	0.866134
O	-4.202994	-2.446771	-1.606364
H	-4.815538	-2.745547	-2.293900
H	-3.524131	-3.146223	-1.512561
O	-4.039096	-1.368186	3.220309
H	-4.347295	-0.697773	3.847243
H	-3.057818	-1.426209	3.334646
O	-1.325917	-1.601147	3.219517
H	-0.861179	-0.773167	3.484502
H	-1.326299	-1.598329	2.244813
O	-1.929467	-3.928335	-0.944325
H	-1.824531	-3.262479	-0.232421
H	-1.962594	-4.794636	-0.510672
O	0.020229	0.745936	3.737360
H	0.867511	0.556952	4.195077
H	0.244010	0.845014	2.792514
O	2.514637	0.043649	4.776656
H	2.785765	0.393054	5.638386
H	3.110294	0.444560	4.118599
O	-0.139068	2.923000	-0.159674
H	-0.111532	2.046062	0.278598
H	0.795359	3.108709	-0.397322
O	-1.926570	2.466129	-2.077847
H	-1.198007	2.763159	-1.466000
H	-1.639822	2.673502	-2.980306
O	-6.521478	2.302234	-1.758473
H	-5.685087	2.815834	-1.648551
H	-6.411417	1.773102	-2.561412
O	-7.039131	0.778562	0.424768
H	-7.231832	1.355717	1.177634
H	-6.905282	1.370774	-0.355950
O	-4.252347	3.735812	-1.316055
H	-3.415142	3.330959	-1.638036
H	-4.284899	4.624087	-1.700033

O	2.272796	-3.091512	-1.559692
H	1.518437	-3.281683	-2.158385
H	2.859295	-2.464646	-2.031676
O	3.954722	-1.145452	-2.739274
H	4.839666	-1.202590	-2.295182
H	4.115493	-1.375974	-3.667990
O	-0.070360	-3.538795	-2.990742
H	-0.063486	-4.225659	-3.673225
H	-0.749423	-3.808953	-2.336387
O	6.380532	-1.027118	-1.519358
H	6.514723	-1.607640	-0.755920
H	6.341763	-0.110753	-1.165902
O	5.658293	1.535574	-0.763158
H	4.794197	1.164934	-0.480162
H	5.443572	1.913820	-1.635358
O	4.419496	3.067045	1.267340
H	5.196554	2.823473	0.727100
H	3.723025	3.265250	0.597104
O	2.567759	3.304949	-0.791474
H	2.861765	2.604503	-1.423118
H	2.670505	4.141290	-1.272671
O	3.645062	1.630538	-2.689148
H	3.625150	0.645641	-2.685701
H	3.459477	1.902432	-3.600275

Table S77. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 19H_2O$ peptide-water complex in the **$19H_2O$_PPII-PPII_B** conformation.

N	-4.661277	0.611444	0.756201
C	-3.671364	-0.075402	-0.108991
C	-2.357459	-0.128357	0.651333
O	-2.096148	0.731673	1.511799
N	-1.493430	-1.074747	0.297413
C	-0.134085	-1.042684	0.806538
C	0.510849	0.295625	0.452333
O	0.310529	0.826964	-0.655192
N	1.341426	0.823733	1.356439
C	2.229513	1.893451	0.939327
C	3.062581	1.381231	-0.229464
O	3.333345	0.190475	-0.343303
O	3.467053	2.298082	-1.077382
C	-3.483201	0.694385	-1.415005
C	0.683248	-2.173641	0.185944
C	3.145794	2.312229	2.088328
H	-4.385653	1.606696	0.916838
H	-4.705401	0.115476	1.663592
H	-5.600842	0.587432	0.299093
H	-4.040338	-1.087472	-0.300996
H	-1.738676	-1.731904	-0.451357
H	-0.149693	-1.155881	1.894859
H	1.660308	0.205900	2.119506
H	1.639772	2.753040	0.605012
H	3.933845	1.853302	-1.875239
H	-4.442691	0.798682	-1.929194
H	-2.802125	0.150386	-2.075066
H	-3.066769	1.688055	-1.217290
H	0.270202	-3.139418	0.490739
H	1.721815	-2.113498	0.521871
H	0.663077	-2.108862	-0.907793

H	2.538906	2.664123	2.926005
H	3.803732	3.123293	1.767684
H	3.757303	1.468077	2.421507
O	-4.031675	-1.304247	2.692858
H	-4.624135	-1.933747	3.128763
H	-3.327172	-1.091043	3.339136
O	-3.967498	3.332346	1.002811
H	-4.325976	3.875975	0.286366
H	-2.986395	3.456008	0.982621
O	-1.237811	3.371496	1.063987
H	-0.840815	3.489027	0.169313
H	-1.232131	2.411059	1.228689
O	-1.713816	-0.412524	3.980308
H	-1.636949	0.204832	3.222820
H	-1.727768	0.131632	4.782479
O	-0.006678	3.536266	-1.394122
H	0.805442	4.066693	-1.243627
H	0.274763	2.602901	-1.367186
O	2.338539	4.853231	-0.670601
H	2.571174	5.711611	-1.054357
H	3.035407	4.232115	-0.943939
O	-0.367180	-0.592169	-2.965303
H	-0.295499	-0.038490	-2.159411
H	0.561560	-0.859006	-3.129997
O	-2.106050	-2.441657	-2.151112
H	-1.405611	-1.882632	-2.583607
H	-1.838461	-3.366808	-2.261326
O	-6.753837	-2.066601	-1.870691
H	-5.920381	-2.039983	-2.398999
H	-6.660865	-2.806094	-1.253127
O	-7.071669	0.310541	-0.604203
H	-7.305654	0.983852	-1.259094
H	-7.009175	-0.546715	-1.093547
O	-4.500591	-1.816145	-3.374813
H	-3.648632	-2.101055	-2.974163
H	-4.560651	-2.271667	-4.227128
O	2.393295	-1.179826	3.059635
H	1.664032	-1.755065	3.377305
H	2.931444	-1.723485	2.449029
O	3.937629	-2.775475	1.228434
H	4.876246	-2.460007	1.278056
H	3.939979	-3.663464	1.619514
O	0.087269	-2.540331	3.805055
H	0.121481	-3.125840	4.575703
H	-0.568899	-1.843215	4.017472
O	6.496819	-1.873881	1.035770
H	6.774185	-1.180725	1.652486
H	6.433187	-1.449552	0.151950
O	5.651821	-0.914826	-1.423375
H	4.882765	-0.502970	-0.977444
H	5.286386	-1.768690	-1.733602
O	4.445527	1.003867	-3.113721
H	5.183470	0.448346	-2.789977
H	3.727513	0.354750	-3.280604
O	2.403128	-0.943211	-2.758199
H	2.407104	-0.548389	-1.865719
H	2.809034	-1.820983	-2.594729
O	3.824289	-3.015897	-1.557841
H	3.746775	-3.012750	-0.575414
H	3.913836	-3.943064	-1.823300

Table S78. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+ \cdot 19H_2O$ peptide-water complex in the **$19H_2O$_PPII-PPII_C** conformation.

N	4.704128	0.475748	0.819324
C	3.694890	-0.067302	-0.122362
C	2.400009	0.694063	0.101030
O	2.159619	1.214891	1.205407
N	1.532596	0.708319	-0.905697
C	0.185841	1.208852	-0.697732
C	-0.480847	0.411380	0.423764
O	-0.259071	-0.804688	0.561988
N	-1.337450	1.067319	1.211369
C	-2.189046	0.291536	2.098643
C	-3.028905	-0.643046	1.235150
O	-3.525840	-0.250816	0.186645
O	-3.195900	-1.857264	1.702995
C	3.474188	-1.556809	0.135348
C	-0.619864	1.069466	-1.986346
C	-3.099886	1.214625	2.903959
H	4.439444	0.269053	1.809108
H	4.762399	1.500001	0.685845
H	5.632955	0.043191	0.614852
H	4.062392	0.104018	-1.138875
H	1.743776	0.189997	-1.765638
H	0.231754	2.263121	-0.408482
H	-1.672535	1.992072	0.896885
H	-1.566165	-0.304410	2.772322
H	-3.695015	-2.429045	1.008870
H	4.419914	-2.096982	0.036332
H	2.770335	-1.962853	-0.596000
H	3.069011	-1.714950	1.140577
H	-0.118260	1.611919	-2.793034
H	-1.617856	1.494745	-1.855046
H	-0.716509	0.016345	-2.273061
H	-2.490145	1.881497	3.518337
H	-3.745676	0.627536	3.561584
H	-3.724602	1.814696	2.235654
O	4.123399	2.994041	-0.261600
H	4.714530	3.608959	-0.719366
H	3.413763	3.535837	0.140940
O	4.024694	-0.252271	3.452622
H	4.368612	-1.121431	3.705001
H	3.042544	-0.297505	3.562996
O	1.301509	-0.157447	3.504013
H	0.886648	-1.028914	3.302216
H	1.304255	0.331608	2.660906
O	1.793531	3.928615	0.987010
H	1.709808	3.002429	1.297533
H	1.824451	4.483856	1.781183
O	0.023424	-2.494001	2.808323
H	-0.772457	-2.538701	3.380552
H	-0.280616	-2.123579	1.958947
O	-2.298464	-2.280725	4.334237
H	-2.546704	-2.940229	4.998712
H	-2.959033	-2.345702	3.623446
O	0.145385	-2.544963	-1.575095
H	0.046321	-1.960555	-0.794767
H	-0.767763	-2.734358	-1.877970
O	1.928955	-1.196250	-3.018806
H	1.197999	-1.757039	-2.640495
H	1.656051	-0.941987	-3.913524
O	6.521460	-1.203198	-2.655896
H	5.685734	-1.713257	-2.784955

H	6.413928	-0.378340	-3.150736
O	7.052469	-0.751886	-0.034910
H	7.300322	-1.586483	0.388173
H	6.909101	-0.955670	-0.991970
O	4.251553	-2.683552	-2.912844
H	3.418917	-2.171233	-3.023726
H	4.289753	-3.298017	-3.660228
O	-2.391479	3.372237	-0.059972
H	-1.648012	3.870190	-0.463527
H	-2.934583	3.031906	-0.800584
O	-3.903104	2.294819	-2.254580
H	-4.849052	2.167109	-1.997029
H	-3.909531	2.962489	-2.957344
O	-0.054458	4.538315	-1.020738
H	-0.072836	5.464816	-1.302034
H	0.613749	4.481070	-0.305299
O	-6.508169	1.695485	-1.613948
H	-6.877047	2.138820	-0.835999
H	-6.533924	0.735696	-1.417238
O	-5.893864	-1.020576	-1.139449
H	-5.300074	-0.692668	-0.437154
H	-5.310743	-0.983607	-1.925765
O	-4.320680	-3.251148	-0.160505
H	-5.096792	-2.769842	-0.509407
H	-3.672998	-3.265016	-0.901764
O	-2.545201	-2.985041	-2.326538
H	-2.860412	-2.145356	-2.726953
H	-2.643662	-3.669605	-3.006200
O	-3.550191	-0.469977	-2.574220
H	-3.280524	-0.297115	-1.651400
H	-3.614776	0.428396	-2.952198

Table S79. M062X/6-31+G*/PCM optimized Cartesian coordinates of the $Ala_3H^+{\cdot}19H_2O$ peptide-water complex in the **$19H_2O$_PPII-PPII_D** conformation.

N	4.751402	0.731639	0.541574
C	3.709843	-0.139163	-0.056151
C	2.431305	0.674036	-0.159086
O	2.226465	1.624912	0.617153
N	1.539112	0.276984	-1.061873
C	0.196596	0.822978	-1.036365
C	-0.449162	0.540874	0.317625
O	-0.156139	-0.473148	0.972876
N	-1.393715	1.398085	0.715988
C	-2.334123	0.972010	1.734133
C	-3.034350	-0.293866	1.242343
O	-3.183204	-0.531663	0.054162
O	-3.496409	-1.064713	2.207317
C	3.477316	-1.367292	0.821725
C	-0.654655	0.186490	-2.134065
C	-3.375789	2.060831	1.988815
H	4.504455	0.989302	1.523897
H	4.832264	1.589062	-0.031688
H	5.665442	0.225683	0.541638
H	4.052733	-0.429743	-1.054048
H	1.723513	-0.563005	-1.621669
H	0.242711	1.905152	-1.190433
H	-1.693440	2.120883	0.051830
H	-1.798114	0.746797	2.661276
H	-3.926264	-1.905329	1.798759

H	4.408666	-1.929767	0.932987
H	2.736153	-2.023466	0.357821
H	3.112924	-1.067487	1.810005
H	-0.203468	0.380097	-3.110984
H	-1.661566	0.609091	-2.114669
H	-0.738771	-0.895191	-1.981693
H	-2.879306	2.970801	2.334548
H	-4.081544	1.733673	2.755854
H	-3.930328	2.283807	1.070880
O	4.199022	2.488407	-1.558489
H	4.797180	2.798723	-2.253525
H	3.524220	3.187267	-1.436709
O	4.137143	1.234211	3.243746
H	4.451996	0.530747	3.829752
H	3.159401	1.296363	3.382739
O	1.426752	1.497006	3.316825
H	0.960813	0.661261	3.553716
H	1.404576	1.541594	2.343462
O	1.939727	3.958003	-0.818816
H	1.855484	3.283772	-0.112084
H	1.986720	4.819223	-0.376271
O	0.104388	-0.882173	3.741382
H	-0.755777	-0.766163	4.199069
H	-0.111692	-0.959159	2.792769
O	-2.455875	-0.414921	4.762118
H	-2.716105	-0.823322	5.600880
H	-3.037870	-0.795432	4.079746
O	0.056164	-2.922625	-0.322077
H	0.048763	-2.086246	0.187909
H	-0.877277	-3.058902	-0.597008
O	1.880252	-2.351032	-2.175598
H	1.142694	-2.680290	-1.591357
H	1.599463	-2.495616	-3.092075
O	6.479054	-2.276821	-1.910525
H	5.635347	-2.779220	-1.805233
H	6.370846	-1.727949	-2.700362
O	7.054274	-0.815916	0.300525
H	7.265591	-1.414326	1.031467
H	6.898156	-1.385816	-0.492520
O	4.191817	-3.689323	-1.487643
H	3.358533	-3.260619	-1.787884
H	4.208673	-4.563347	-1.903993
O	-2.241756	3.119439	-1.443417
H	-1.470236	3.367426	-2.011803
H	-2.669286	3.953427	-1.195214
O	-4.073138	1.317429	-2.632143
H	-4.938747	1.386538	-2.172414
H	-3.465910	1.944457	-2.188431
O	0.046847	3.588110	-2.829788
H	0.043594	4.249927	-3.536891
H	0.744678	3.860427	-2.195398
O	-6.573639	1.121801	-1.422640
H	-6.741166	1.636969	-0.620356
H	-6.450162	0.192376	-1.130247
O	-5.653392	-1.432460	-0.866552
H	-4.795469	-1.067563	-0.560787
H	-5.433518	-1.732218	-1.768176
O	-4.446861	-3.147735	1.019301
H	-5.213339	-2.831616	0.500990
H	-3.751592	-3.285574	0.333204
O	-2.619269	-3.210301	-1.078453
H	-2.906726	-2.447743	-1.638357
H	-2.722188	-3.994660	-1.640116
O	-3.674458	-1.357747	-2.811731

H	-3.699308	-0.369475	-2.723159
H	-3.488029	-1.543871	-3.743579