

Table S1. α -Glycine ADPs (\AA^2)

neutron*		U11	U22	U33	U12	U13	U23
	H1	0.01873	0.02257	0.01248	0.00162	0.00755	0.00185
	H2	0.01177	0.02399	0.01769	-0.0014	0.00371	-0.0029
	H3	0.02460	0.01243	0.02274	0.00321	0.00984	0.00186
	H4	0.02846	0.01190	0.02428	0.00306	0.01380	0.00453
	H5	0.01180	0.03156	0.01591	-0.0020	0.00202	-0.0046
SHADE2	H1	0.02262	0.02814	0.00965	0.00407	0.00867	0.00299
	H2	0.01027	0.02226	0.01731	-0.00291	0.00266	-0.00369
	H3	0.02203	0.00977	0.02331	0.00104	0.01068	0.00084
	H4	0.03149	0.01227	0.02805	0.00235	0.01663	0.00468
	H5	0.01346	0.03416	0.01636	-0.00305	0.00164	-0.00586
SHADE3	H1	0.02122	0.02812	0.01290	0.00429	0.00942	0.00213
	H2	0.01172	0.01852	0.01793	-0.00124	0.00407	-0.00187
	H3	0.01881	0.01182	0.02187	0.00037	0.00796	-0.00106
	H4	0.02003	0.01168	0.02365	0.00119	0.01023	0.00414
	H5	0.01173	0.02256	0.01429	-0.00036	0.00126	-0.00344

Table S2. 1-Methyluracil ADPs (Å²)

neutron		U11	U22	U33	U12	U13	U23
	H3	0.0151	0.0104	0.0245	-0.0028	0.0000	0.0000
	H5	0.0193	0.0115	0.0331	0.0034	0.0000	0.0000
	H6	0.0155	0.0118	0.0275	-0.0050	0.0000	0.0000
	H11	0.0170	0.0152	0.0544	-0.0055	0.0000	0.0000
	H12	0.0221	0.0398	0.0311	-0.0033	-0.0067	0.0141
SHADE2		U11	U22	U33	U12	U13	U23
	H3	0.0164	0.0111	0.0367	-0.0039	0.0000	0.0000
	H5	0.0189	0.0104	0.0328	0.0042	0.0000	0.0000
	H6	0.0169	0.0125	0.0387	-0.0057	0.0000	0.0000
	H11	0.0214	0.0144	0.0553	-0.0052	0.0000	0.0000
	H12	0.0225	0.0366	0.0275	0.0037	-0.0045	0.0124
SHADE3		U11	U22	U33	U12	U13	U23
	H3	0.015179	0.010676	0.029208	-0.00318	0.000000	0.000000
	H5	0.017875	0.010865	0.031094	0.003810	0.000000	0.000000
	H6	0.015531	0.011853	0.033607	-0.00405	0.000000	0.000000
	H11	0.016647	0.013726	0.047867	-0.00413	0.000000	0.000000
	H12	0.019381	0.032971	0.024871	0.004395	-0.00388	0.010102
SHADE3 Xray		U11	U22	U33	U12	U13	U23
	H3	0.015519	0.010516	0.027278	-0.00357	0.000000	0.000000
	H5	0.018435	0.010675	0.032084	0.003820	0.000000	0.000000
	H6	0.015991	0.011793	0.032207	-0.00457	0.000000	0.000000
	H11	0.016757	0.014516	0.046647	-0.00465	0.000000	0.000000
	H12	0.019401	0.033571	0.025761	0.004175	-0.00395	0.010562

Similarity_index	Neutron		neutron_scaled
	SHADE2	SHADE3	SHADE3 Xray
H3	0.47	0.20	0.07
H5	0.20	0.13	0.17
H6	0.39	0.34	0.16
H11	0.58	0.22	0.26
H12	0.22	0.63	0.29
Mean	0.37	0.30	0.19

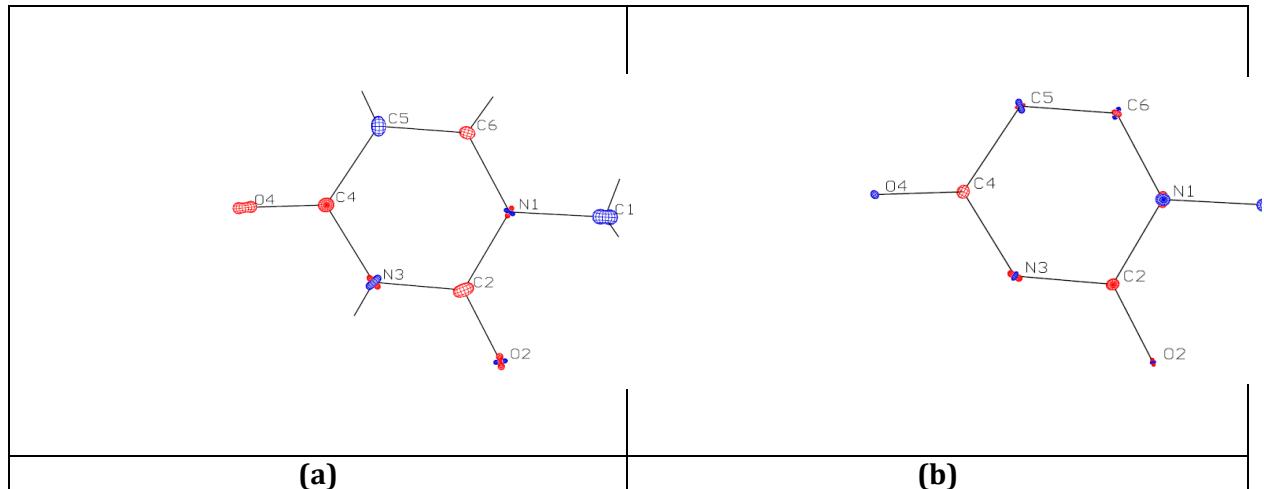


Fig. 1 Difference between U_{ij} observed and U_{ij} calculated from TLS analysis - (a) after neutron measurement (b)after SHADE3

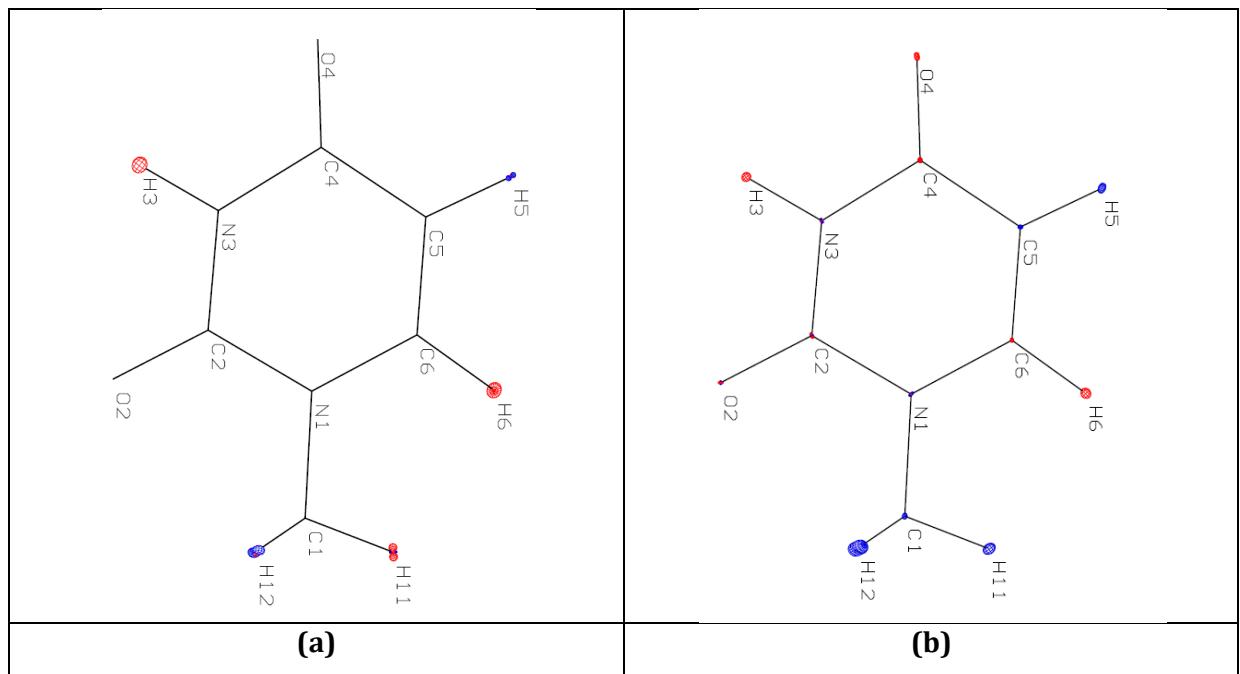


Table S3. L-Alanine(Å²)

60K		U11	U22	U33	U12	U13	U23
neutron	H1	0.0226	0.0186	0.0272	0.005	0.0037	0.0013
	H2	0.0249	0.0139	0.0209	0.0045	0.0006	0.0024
	H3	0.0319	0.0161	0.0375	-0.0015	-0.0035	0.0009
	H4	0.0214	0.0244	0.0134	0.0011	-0.0007	0.0001
	H5	0.0187	0.0348	0.0273	-0.0023	0.0064	-0.0041
	H6	0.0164	0.0256	0.0228	-0.0052	0.0006	-0.0021
	H7	0.0265	0.0366	0.0197	-0.0046	-0.009	0.0017
SHADE2	H1	0.0256	0.0158	0.0232	0.0069	0.0020	0.0001
	H2	0.0260	0.0131	0.0225	0.0044	-0.0010	0.0013
	H3	0.0332	0.0176	0.0470	-0.0020	-0.0047	-0.0014
	H4	0.0237	0.0291	0.0113	0.0019	-0.0030	0.0014
	H5	0.0230	0.0482	0.0285	-0.0061	0.0101	-0.0085
	H6	0.0179	0.0242	0.0230	-0.0076	0.0007	-0.0014
	H7	0.0329	0.0446	0.0202	-0.0081	-0.0108	0.0068
SHADE3	H1	0.0247	0.0179	0.0254	0.0070	0.0024	0.0005
	H2	0.0247	0.0131	0.0205	0.0043	-0.0006	0.0006
	H3	0.0316	0.0164	0.0373	-0.0031	-0.0031	-0.0008
	H4	0.0231	0.0265	0.0124	0.0010	-0.0011	0.0007
	H5	0.0199	0.0423	0.0247	-0.0042	0.0078	-0.0040
	H6	0.0184	0.0258	0.0224	-0.0070	0.0014	-0.0008
	H7	0.0267	0.0369	0.0183	-0.0044	-0.0083	0.0030
RT		U11	U22	U33	U12	U13	U23
neutron	H1	0.0430	0.0402	0.044	0.0072	0.0060	0.0019
	H2	0.0540	0.0304	0.0360	0.0106	-0.0021	0.0023
	H3	0.0630	0.0440	0.0710	-0.0140	-0.0020	-0.0050
	H4	0.0470	0.0500	0.0216	0.0024	0.0026	0.0016
	H5	0.0450	0.0960	0.0630	-0.0140	0.0140	-0.0080
	H6	0.0420	0.0570	0.0380	-0.0090	-0.0008	-0.0011
	H7	0.0430	0.0870	0.0410	-0.0110	-0.0120	-0.0030
SHADE2	H1	0.0559	0.0369	0.0409	0.0133	0.0031	-0.0040
	H2	0.0547	0.0301	0.0391	0.0109	-0.0018	0.0037
	H3	0.0731	0.0458	0.0749	-0.0172	-0.0060	-0.0061
	H4	0.0523	0.0596	0.0236	0.0047	-0.0024	0.0018
	H5	0.0458	0.0989	0.0525	-0.0137	0.0106	-0.0081
	H6	0.0414	0.0506	0.0412	-0.0096	0.0026	0.0009
	H7	0.0590	0.0962	0.0403	-0.0135	-0.0167	0.0022
SHADE3	H1	0.0559	0.0396	0.0456	0.0142	0.0039	-0.0028
	H2	0.0544	0.0301	0.0377	0.0112	-0.0015	0.0027
	H3	0.0749	0.0444	0.0775	-0.0182	-0.0060	-0.0053
	H4	0.0517	0.0566	0.0249	0.0034	-0.0005	0.0008
	H5	0.0454	0.1004	0.0528	-0.0144	0.0117	-0.0069
	H6	0.0424	0.0536	0.0412	-0.0096	0.0033	0.0010
	H7	0.0570	0.0940	0.0404	-0.0135	-0.0170	0.0010

Similarity_index 60K	Neutron	
	SHADE2	SHADE3
H1	0.71	0.25
H2	0.16	0.16
H3	0.47	0.14
H4	0.83	0.14
H5	1.06	0.46
H6	0.28	0.20
H7	0.80	0.09
Mean	0.62	0.21

Similarity_index RT	Neutron	
	SHADE2	SHADE3
H1	1.06	0.83
H2	0.06	0.02
H3	0.20	0.27
H4	0.61	0.41
H5	0.24	0.21
H6	0.28	0.24
H7	0.80	0.60
Mean	0.46	0.37

Similarity_index 100K	Neutron_scaled
	SHADE3
H1	0.78
H2	0.29
H3	0.77
H4	0.79
H5	1.23
H6	1.02
H7	1.52
Mean	0.91

Similarity_index 60 K	Neutron		
	SHADE 2	SHADE 3(6-31G(d,p)	SHADE3 (STO-3G)
H1	0.71	0.25	0.74
H2	0.16	0.16	0.19
H3	0.47	0.14	1.18
H4	0.83	0.14	0.4
H5	1.06	0.46	0.86
H6	0.28	0.20	0.74
H7	0.80	0.09	0.99
Mean	0.62	0.21	0.73

Table S4. 2-Methyl-4-nitouracil (Å²)

		U11	U22	U33	U12	U13	U23
Neutron_Scaled	HNA	0.0255	0.0246	0.0398	-0.0019	-0.0069	0.0018
	HNB	0.0306	0.0195	0.0495	0.0033	-0.0084	0.0051
	H3	0.0259	0.0153	0.0395	0.0030	-0.0081	0.0009
	H5	0.0205	0.0196	0.0412	-0.0040	-0.0072	0.0016
	H6	0.0285	0.0179	0.0455	0.0009	-0.0025	0.0033
	H7A	0.0375	0.0376	0.0256	-0.0078	-0.0084	0.0012
	H7B	0.0244	0.0446	0.0406	-0.0082	0.0050	-0.0033
	H7C	0.0353	0.0194	0.0598	0.0001	-0.0131	0.0024
SHADE2	HNA	0.0216	0.0291	0.0398	-0.0042	-0.0086	0.0010
	HNB	0.0288	0.0216	0.0412	0.0027	-0.0064	0.0057
	H3	0.0295	0.0159	0.0370	0.0019	-0.0043	0.0021
	H5	0.0241	0.0234	0.0474	-0.0071	-0.0105	-0.0005
	H6	0.0303	0.0159	0.0452	0.0019	-0.0061	0.0027
	H7A	0.0430	0.0427	0.0274	-0.0106	-0.0124	0.0034
	H7B	0.0232	0.0447	0.0495	-0.0072	0.0088	-0.0060
	H7C	0.0364	0.0203	0.0559	-0.0003	-0.0105	0.0023
SHADE3	HNA	0.0208	0.0293	0.0429	-0.0037	-0.0099	0.0010
	HNB	0.0294	0.0218	0.0470	0.0029	-0.0083	0.0079
	H3	0.0276	0.0164	0.0344	0.0017	-0.0039	0.0022
	H5	0.0233	0.0234	0.0448	-0.0064	-0.0096	-0.0003
	H6	0.0281	0.0161	0.0432	0.0014	-0.0059	0.0022
	H7A	0.0401	0.0393	0.0266	-0.0089	-0.0120	0.0014
	H7B	0.0208	0.0400	0.0486	-0.0058	0.0071	-0.0034
	H7C	0.0343	0.0188	0.0488	-0.0022	-0.0082	0.0027

Similarity_Index		NEUTRON
	SHADE2	SHADE3
HNA	0.64	0.90
HNB	0.33	0.20
H3	0.47	0.50
H5	0.69	0.48
H6	0.27	0.25
H7A	0.31	0.18
H7B	0.39	0.50
H7C	0.09	0.45
Mean	0.40	0.43

Table S5. Xylitol (Å²)

		U11	U22	U33	U12	U13	U23
neutron	H1A	0.0321	0.0266	0.0249	0.0081	-0.0033	-0.0117
	H1B	0.0181	0.0383	0.0272	-0.0067	0.0025	0.0117
	H2	0.0207	0.0187	0.0226	0.0008	0.0012	-0.0072
	H3	0.0222	0.0184	0.019	0.0017	-0.0011	-0.0067
	H4	0.0232	0.0271	0.0136	-0.0005	-0.0009	0.0047
	H5B	0.0237	0.0317	0.0272	-0.0006	-0.0046	0.0122
	H5A	0.0234	0.0254	0.0444	0.0021	0.0035	-0.018
	H11	0.0163	0.0299	0.0187	-0.0011	0.0037	0.0032
	H12	0.0256	0.0286	0.0195	-0.0046	0.0061	-0.0015
	H13	0.0301	0.0157	0.0212	-0.0006	0.0007	0.0023
	H14	0.0307	0.0179	0.0256	0.0085	-0.0037	0.002
	H15	0.0245	0.0264	0.0203	-0.0061	0	-0.0077
SHADE2	H1A	0.0289	0.0236	0.0224	0.0031	-0.0029	-0.0083
	H1B	0.0205	0.0295	0.0243	-0.0063	0.0008	0.0070
	H2	0.0224	0.0185	0.0192	0.0025	0.0015	-0.0057
	H3	0.0234	0.0188	0.0200	0.0024	0.0011	-0.0069
	H4	0.0230	0.0255	0.0137	-0.0005	0.0003	0.0036
	H5B	0.0246	0.0325	0.0183	0.0004	-0.0030	0.0042
	H5A	0.0236	0.0266	0.0284	0.0023	0.0016	-0.0124
	H11	0.0171	0.0298	0.0177	0.0021	0.0029	0.0012
	H12	0.0231	0.0283	0.0150	-0.0044	0.0044	-0.0013
	H13	0.0292	0.0159	0.0227	-0.0016	-0.0026	0.0022
	H14	0.0247	0.0173	0.0214	0.0037	-0.0035	0.0010
	H15	0.0219	0.0269	0.0205	-0.0045	-0.0017	-0.0044

SHADE3	H1A	0.0241	0.0220	0.0199	0.0023	-0.0010	-0.0077
	H1B	0.0176	0.0255	0.0205	-0.0046	0.0014	0.0048
	H2	0.0216	0.0176	0.0179	0.0018	0.0011	-0.0048
	H3	0.0218	0.0192	0.0188	0.0022	0.0005	-0.0067
	H4	0.0213	0.0255	0.0133	-0.0008	0.0006	0.0030
	H5B	0.0233	0.0295	0.0194	0.0001	-0.0025	0.0060
	H5A	0.0215	0.0232	0.0308	0.0020	0.0015	-0.0117
	H11	0.0172	0.0294	0.0184	0.0008	0.0036	0.0007
	H12	0.0283	0.0308	0.0154	-0.0073	0.0070	-0.0017
	H13	0.0308	0.0150	0.0235	-0.0014	-0.0025	0.0026
	H14	0.0300	0.0177	0.0240	0.0060	-0.0034	0.0020
	H15	0.0246	0.0273	0.0204	-0.0064	-0.0019	-0.0054
	H12	0.0282	0.0308	0.0152	-0.0068	0.0069	-0.0012
	H13	0.0304	0.0151	0.0234	-0.0011	-0.0019	0.0027
	H14	0.0294	0.0176	0.0234	0.0058	-0.0033	0.0012
	H15	0.0246	0.0277	0.0202	-0.0064	-0.0020	-0.0056

Similarity_index	NEUTRON	
	SHADE2	SHADE3
H1A	0.63	1.22
H1B	0.84	1.57
H5A	1.44	0.93
H5B	1.66	0.96
H11	0.48	0.33
H12	0.48	0.65
H13	0.26	0.26
H14	0.95	0.20
H15	0.50	0.24
H2	0.31	0.42
H3	0.23	0.13
H4	0.11	0.25
Mean	0.66	0.60

Figure S3. Proton sponges complexes used for analysis: (a) -1 (b) - 2 (c) - 3 (d) - 4.

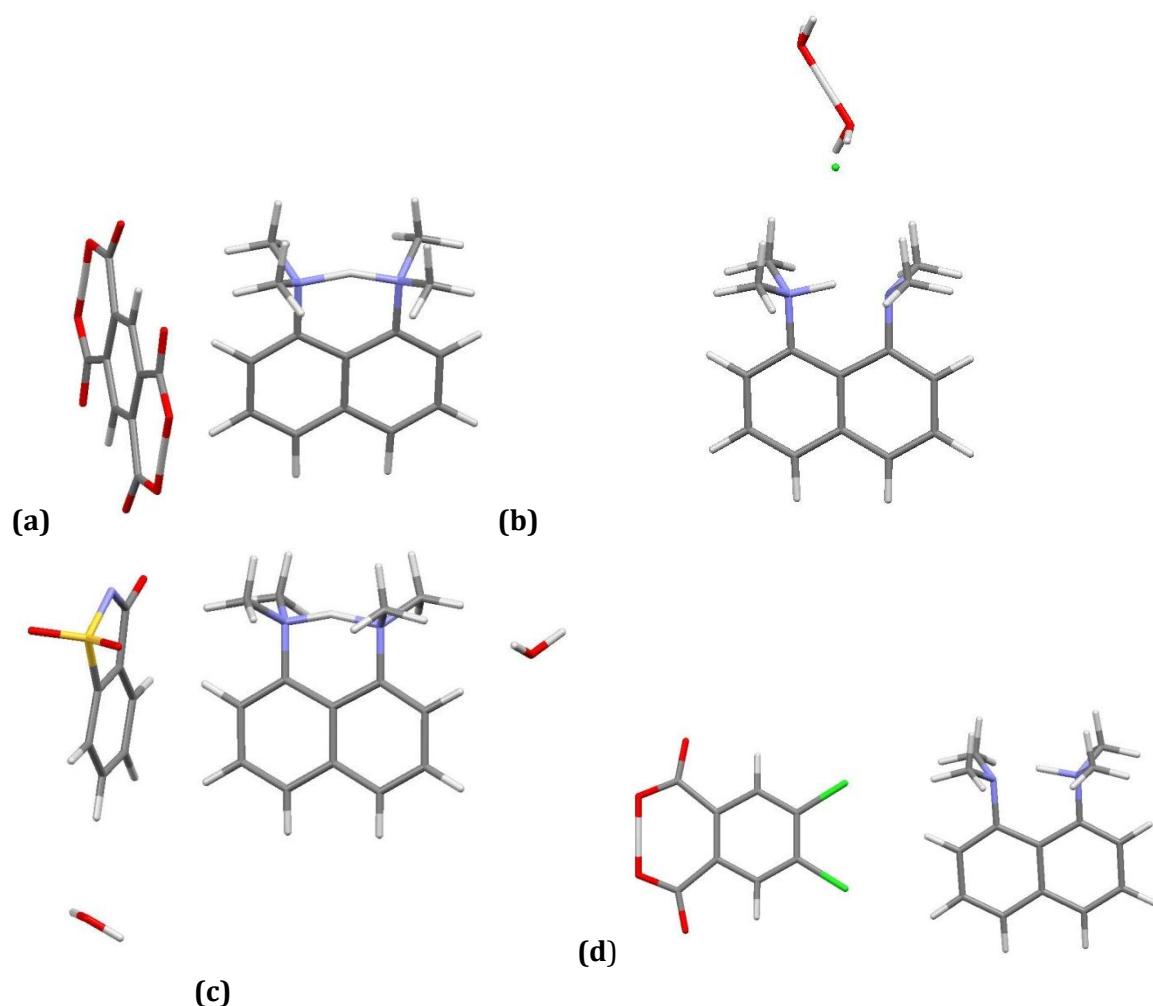


Table S6. Internal mean square displacements derived for H1N from N1-H1N...N2 hydrogen bond from proton sponges.

Proton sponge id	msd bond direction	msd in-plane	msd out-of plane
1	283	219	153
2	245	101	130
3	250	44	90
4	419	125	175