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

Supplementary data

Entropy connects water structure and dynamics in protein hydration layer

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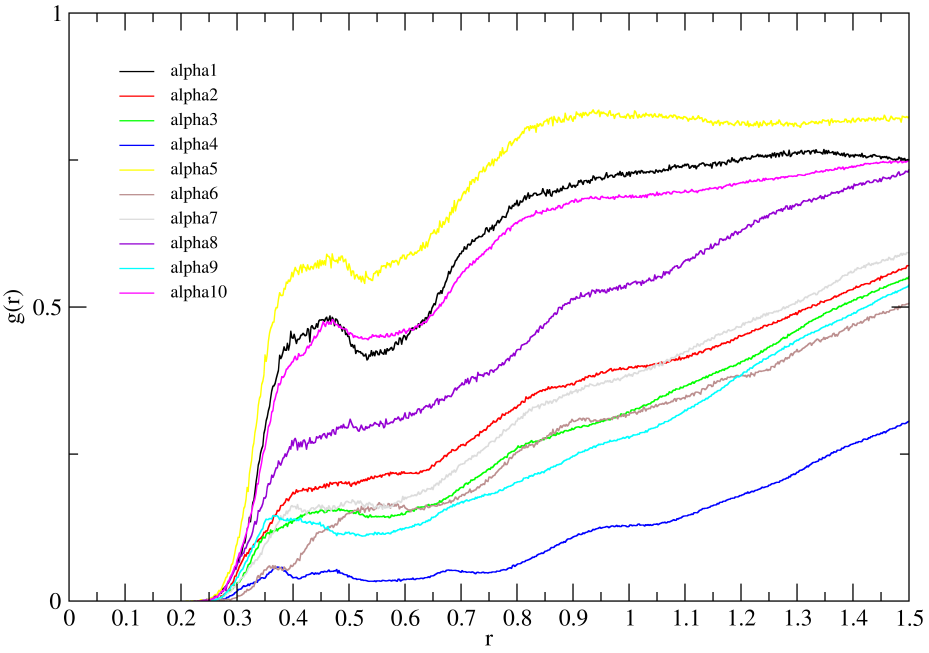

Figure S1. Solvent radial distribution functions were also calculated for water molecules around the C_α atoms of the protein backbone.

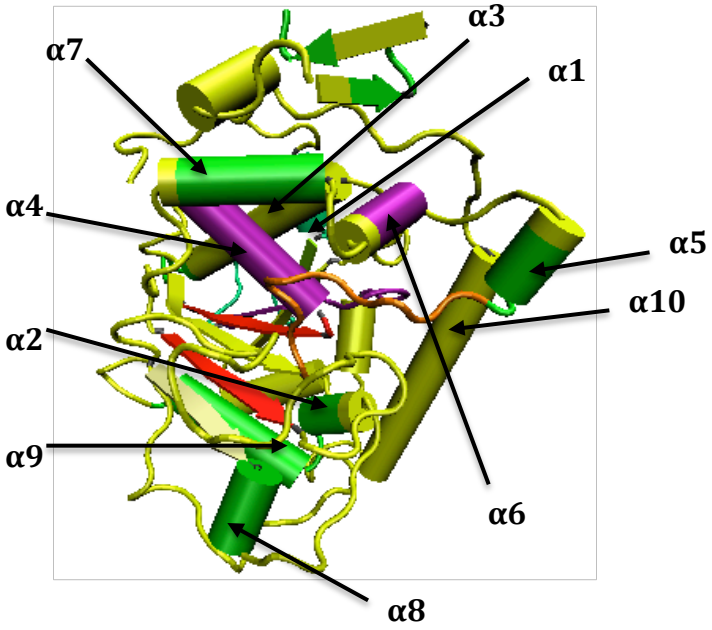

Figure S2. CALB structure color coded according to water-protein hydrogen bond life times in GROMOS 53a6 forcefield. Green <30 ps, yellow 30-75 ps, orange 75-100 ps, pink 100-1000 ps, and red >1000 ps.

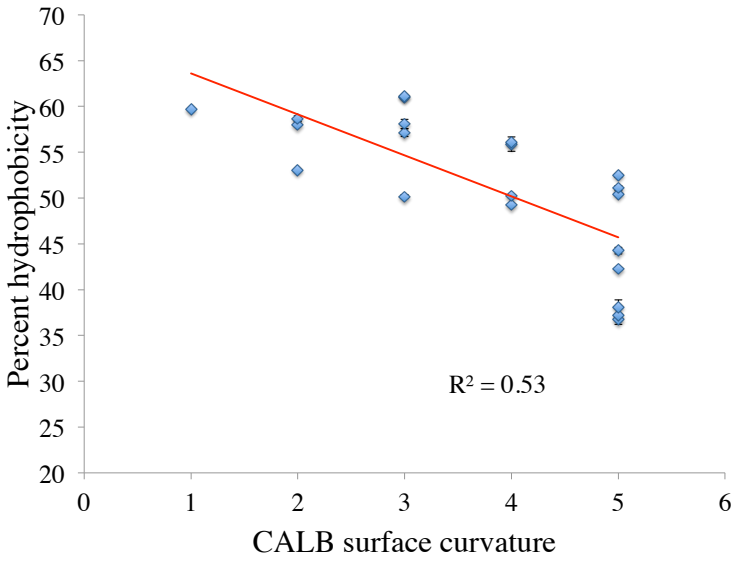

Figure S3. Correlation between CALB surface percent hydrophobicity and CALB surface curvature.

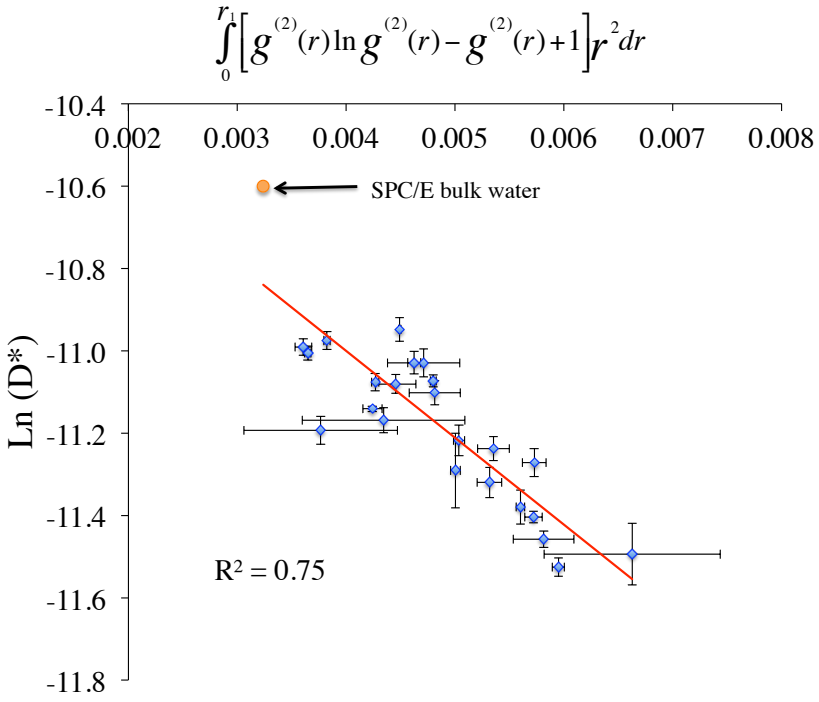

Figure S4. Logarithmic relationship between hydration shell water diffusivity and integral part of the pair entropy calculation, as described in Eq. 9 with SPC/E water model. A data point for the SPC/E bulk water is also shown here. The correlation coefficient contains the data point for the SPC/E bulk water.

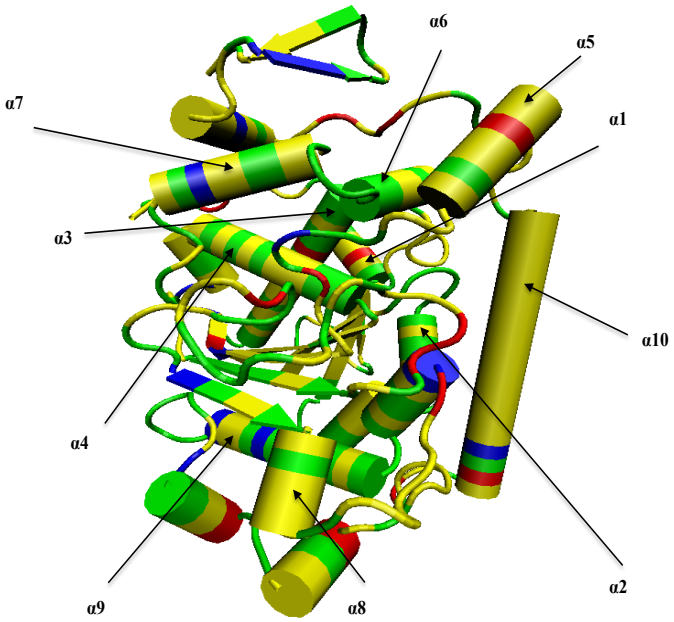

Figure S5. CALB surface color-coded by amino acid type. Yellow indicates hydrophobic residues, green indicates polar residues, red indicates acidic residues, and blue corresponds to basic residues.

Table S1. Interfacial hydration dynamics around helix α5 in different conformations**.**

Conformation	Hydrogen bond lifetime (ps)	Residence time (ps)	Reorientation time (ps)
Crystal-like	24.4 ± 3.4	39.8 ± 1.2	2.42 ± 0.04
Close	26.6 ± 1.0	43.1 ± 0.6	2.40 ± 0.02
Open	20.8 ± 1.1	40.7 ± 1.0	2.39 ± 0.04

Table S2. Reorientation times (τ_2) calculated for the water O-H vectors.

	Reorientation time (ps)
α1	2.3 ± 0.1
α2	3.4 ± 0.2
α3	3.7 ± 0.2
α4	6.0 ± 0.5
α5	2.7 ± 0.3
α6	5.8 ± 1.0
α7	2.6 ± 0.1
α8	2.6 ± 0.1
α9	3.5 ± 0.5
α10	2.9 ± 0.1

Table S3. Water-protein hydrogen bond lifetimes (HBLTs) obtained by fitting autocorrelation functions with both biexponential and triexpoential fits ($\Sigma A_i e^{-t/\tau_i}$), where values of HBLT=$\Sigma A_i \tau_i$.

	HBLT (ps) with biexponential fit	**HBLT (ps) with triexponential fit**
α1	44.5 ± 9.8	38.2 ± 10.1
α2	196 ± 32	198 ± 35
α3	230 ± 20	243 ± 17
α4	315 ± 88	315 ± 88
α5	49.8 ± 7.5	47.9 ± 16.2
α6	763 ± 144	825 ± 46
α7	60.7 ± 17.6	62.2 ± 11.4
α8	36.6 ± 1.6	36.6 ± 2.4
α9	154 ± 32	160 ± 51
α10	158 ± 21	187 ± 15

Table S4. Some properties of solvent exposed secondary structures.

Solvent exposed secondary structures	HBLT (order of increas-ing)	Number of h-bonding side chains	Number of doubly h-bonding side chains	% 1-4 h-bond donor neigh-bours	% surface area with h-bonding side chains	% surface area with doubly h-bonding side chains	Number of hydro-phobic residues	Number of acidic residues	Number of basic residues
α1	22.76 ± 0.40	3	2	33%	65%	35%	3	1	1
β7	23.07 ± 3.20	3	2	33%	94%	42%	1	0	1
α5	23.80 ± 1.70	1	1	0%	4%	4%	4	1	0
α8	25.08 ± 0.81	1	1	0%	13%	13%	3	0	0
β1	25.46 ± 1.29	1	1	-	73%	73%	1	0	0
β8	28.01 ± 1.48	2	2	-	100%	100%	0	0	1
β9	30.01 ± 4.12	0	0	-	-	-	2	0	0
α10	40.06 ± 2.10	3	2	0%	21%	15%	17	1	1