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Dynamic coupling of hydration layer to a fluid phospholipid membrane: Intermittency and multiple time-scale relaxations

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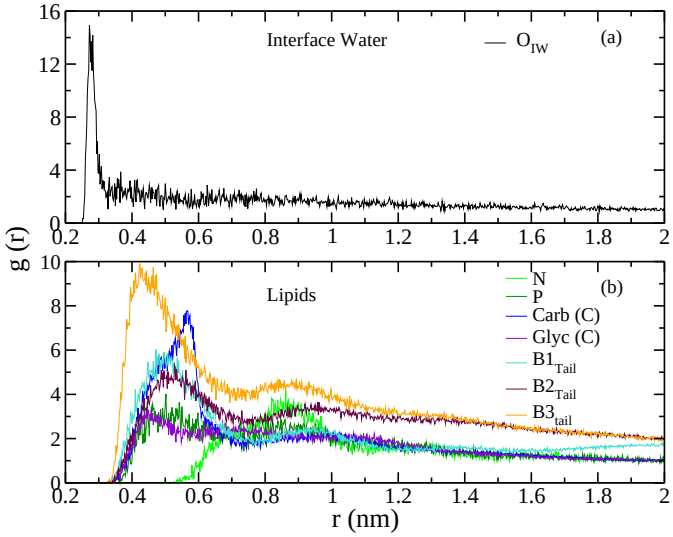

Figure S1 RDF of a) oxygen-oxygen of IW, of b) lipid components from head to tail along the chain.

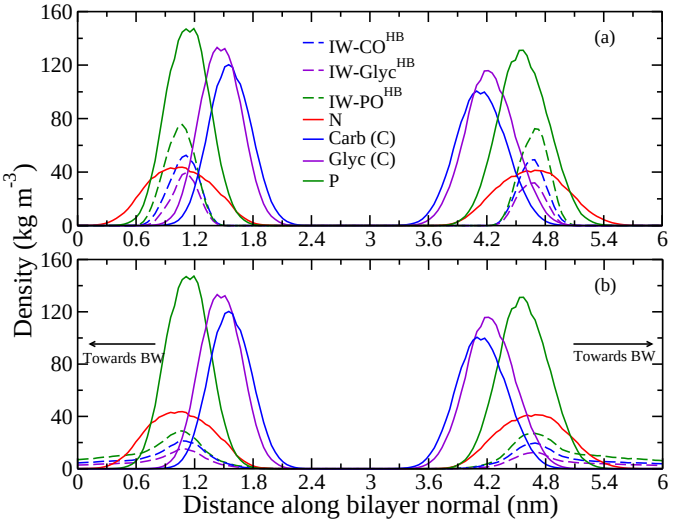

Figure S2 Density profiles of different lipid head moieties and IW^{HB} hydrogen bonded to those moieties along the bilayer normal. The profiles are averaged over 10 sets of a) 100 ps and b) 1 ns runlengths. Different classes of IW^{HB} are confined close to the peak location of N and form hydrogen bonds to oxygens of P, Carb, Glyc. With time, the IW^{HB} molecules diffuse towards bulk water (BW) region.

Table S1 Time-scales when IW^{HB} molecules enter sub-diffusive regime for 10 independent sets each with 1 ns run length.

Beads	t_1 (ps)	t_2 (ps)	t_3 (ps)	t_4 (ps)	t_5 (ps)	t_6 (ps)	t_7 (ps)	t_8 (ps)	t_9 (ps)	t_{10} (ps)
IW^{HB}	6.07	5.11	5.75	6.51	5.02	5.08	7.86	5.51	6.58	5.37
IW-CO^{HB}	6.94	5.93	6.22	7.06	5.71	5.56	8.56	5.68	6.70	6.03
IW-$Glyc^{HB}$	6.26	5.22	6.05	8.11	5.60	5.89	10.70	5.87	6.74	6.11
IW-PO^{HB}	6.14	5.06	5.74	6.46	3.73	5.10	7.44	5.45	6.34	5.31

Table S2 Time-scales when IW^{d} molecules enter sub-diffusive regime for 10 independent sets each with 1 ns run length.

Beads	t_1 (ps)	t_2 (ps)	t_3 (ps)	t_4 (ps)	t_5 (ps)	t_6 (ps)	t_7 (ps)	t_8 (ps)	t_9 (ps)	t_{10} (ps)
IW-CO^{d}	12.61	10.90	11.62	12.98	11.78	9.69	10.63	13.36	12.42	12.83
IW-$Glyc^{d}$	12.91	10.77	11.17	12.51	8.76	12.16	8.18	11.82	13.49	10.80
IW-PO^{d}	7.87	7.38	7.24	6.01	7.30	6.36	6.07	7.18	6.86	7.24
IW-N^{d}	6.51	5.78	5.45	4.96	6.37	5.71	5.32	6.00	5.48	5.91

Table S3 Time-scales when DMPC beads enter sub-diffusive regime for 10 independent sets each with 1 ns run length.

Beads	t_1 (ps)	t_2 (ps)	t_3 (ps)	t_4 (ps)	t_5 (ps)	t_6 (ps)	t_7 (ps)	t_8 (ps)	t_9 (ps)	t_{10} (ps)
$Carb_{Head}$	169.29	85.70	199.22	156.31	175.01	196.71	154.36	187.35	147.32	91.92
$Glyc_{Head}$	180.70	97.43	218.87	165.34	289.77	217.72	177.55	199.95	164.09	302.62
P_{Head}	118.60	73.77	162.12	127.27	134.01	147.51	131.88	126.11	107.99	189.72
N_{Head}	73.04	89.92	83.45	83.44	77.28	88.29	81.30	80.22	69.70	86.81
$B1_{Tail}$	73.19	87.14	77.66	77.66	71.82	83.90	71.04	84.10	73.52	93.25
$B2_{Tail}$	18.93	19.38	18.65	18.65	19.59	19.26	18.79	19.02	19.63	18.01
$B3_{Tail}$	2.97	3.11	2.86	2.86	2.93	2.82	2.84	2.96	2.90	3.07

Table S4 Fitting parameters of SISFs of all classes of IW^{HB} and BW calculated at $\lambda = 0.6$ nm. Correlation coefficients were >0.99.

Region	τ_s (ps)	f_Q	τ_α (ps)	β_α	$f_{Q'}$	τ_l (ps)	β_l
IW^{N}	0.24	0.50	3.49	0.91	0.45	43.00	0.62
IW-PO^{HB}	0.21	0.41	3.40	0.93	0.58	24.42	0.51
IW-CO^{HB}	0.25	0.36	4.03	0.94	0.59	31.54	0.53
IW-$Glyc^{HB}$	0.22	0.33	3.90	0.99	0.64	30.64	0.54
BW	0.10	0.90	3.54	0.94			

Table S5 Fitting parameters of SISFs of all classes of IW^{d} calculated at $\lambda = 0.6$ nm. Correlation coefficients were >0.99.

Region	τ_s (ps)	f_Q	τ_α (ps)	β_α	$f_{Q'}$	τ_l (ps)	β_l
IW-N^{d}	0.2	0.53	3.59	0.89	0.42	44	0.64
IW-PO^{d}	0.28	0.52	5.0	0.76	0.41	62	0.64
IW-$Glyc^{d}$	0.54	0.59	16.0	0.47	0.361	100	0.64
IW-CO^{d}	0.84	0.63	21.14	0.47	0.33	106.04	0.63

Table S6 Fitting parameters of SISFs of all classes of IW^{HB} at $\lambda = 0.3$ nm. Correlation coefficients were >0.99.

Region	τ_s (ps)	f_Q	τ_α (ps)	β_α	$f_{Q'}$	τ_l (ps)	β_l
IW^{HB}	0.16	0.46	1.07	0.90	0.31	7.92	0.55
IW-PO^{HB}	0.16	0.50	0.97	0.85	0.30	8.02	0.51
IW-CO^{HB}	0.19	0.42	0.90	0.80	0.36	8.00	0.54
IW-$Glyc^{HB}$	0.18	0.42	0.91	0.82	0.36	8.23	0.54

Table S7 Fitting parameters of SISFs of all classes of IW^d calculated at $\lambda = 0.3$ nm. The fittings of τ_s are comparatively poor to the ones at higher λ to obtain physically meaningful parameters. Correlation coefficients were >0.99.

Region	τ_s (ps)	f_Q	τ_α (ps)	β_α	$f_{Q'}$	τ_l (ps)	β_l
IW-N^d	0.20	0.52	1.097	0.74	0.23	11.47	0.62
IW-PO^d	0.22	0.51	1.50	0.66	0.235	16.09	0.62
IW-$Glyc^d$	0.27	0.63	2.419	0.456	0.18	27.82	0.62
IW-CO^d	0.30	0.80	3.90	0.41	0.06	29.5	0.62

Table S8 Scaling factors for fig. 13 (a) -(i)

	Region	DMPC	$\lambda = 0.80$ nm		$\lambda = 0.60$ nm		$\lambda = 0.30$ nm	
			IW^{HB}	IW^d	IW^{HB}	IW^d	IW^{HB}	IW^d
(a)-(c)	N	1.00	1.00	1.00	1.14	1.00	1.00	1.00
τ_s	P	1.08	1.10	2.21	1.10	1.40	1.10	1.10
	Glyc	1.31	1.10	3.04	1.04	2.70	1.25	1.35
	Carb	1.36	1.02	5.05	1.19	4.20	1.18	1.50
(d)-(f)	N	1.00	1.00	1.00	1.02	1.00	1.18	1.00
τ_α	P	1.33	1.41	1.41	1.00	1.39	1.07	1.37
	Glyc	1.93	1.33	3.02	1.14	4.45	1.01	2.21
	Carb	2.04	1.24	7.71	1.18	5.96	1.00	3.57
(g)-(i)	N	1.00	1.00	1.00	1.76	1.00	1.18	1.00
τ_α(DMPC)	P	1.33	1.04	1.40	1.00	1.40	1.01	1.40
τ_l(IW)	Glyc	1.93	1.20	2.40	1.25	2.27	1.03	1.42
	Carb	2.04	1.07	2.88	1.29	2.40	1.01	2.57

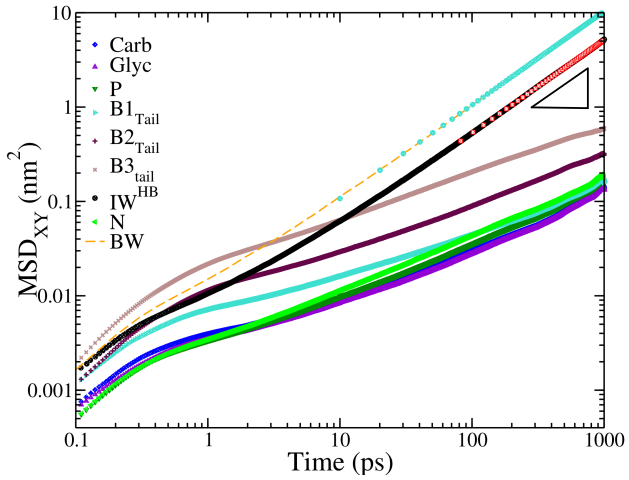

Figure S3 Two dimensional translational mean square displacement (MSD_{XY}) of all beads of DMPC, IW^{HB} and BW.

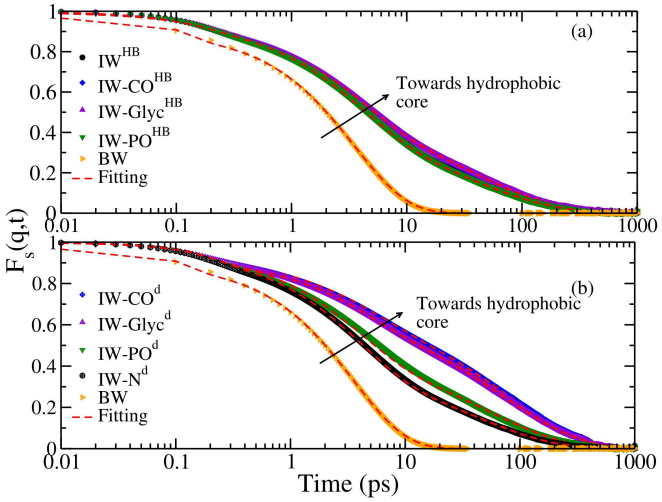

Figure S5 SISF for all classes of a) IW^{HB}, b) IW^{d} at $\lambda = 0.60$ nm. IW-CO^{d} molecules buried deep in the hydrophobic core relax slowest.

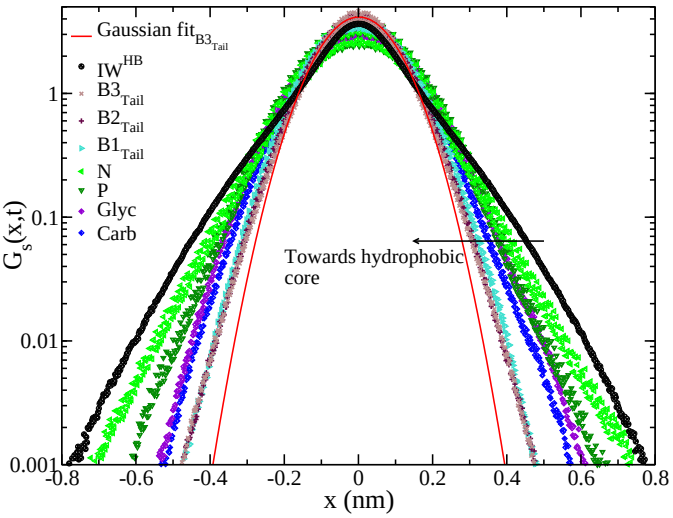

Figure S4 One dimensional van Hove correlation function of DMPC beads and IW^{HB}. End beads of lipid tails have minimum deviations from Gaussianity.

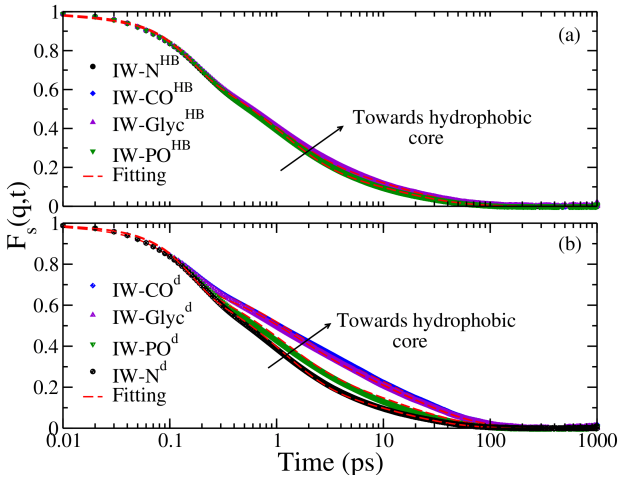

Figure S6 SISF for all classes of a) IW^{HB}, b) IW^{d} at $\lambda = 0.30$ nm. IW-CO^{d} molecules buried deep in the hydrophobic core relax slowest.