Supplementary Information

Relaxation dynamics of saturated and unsaturated oriented lipid bilayers

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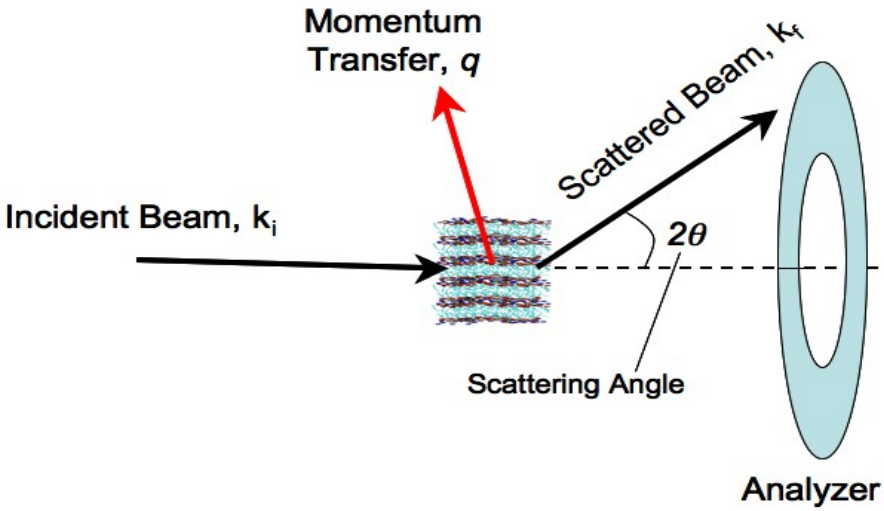
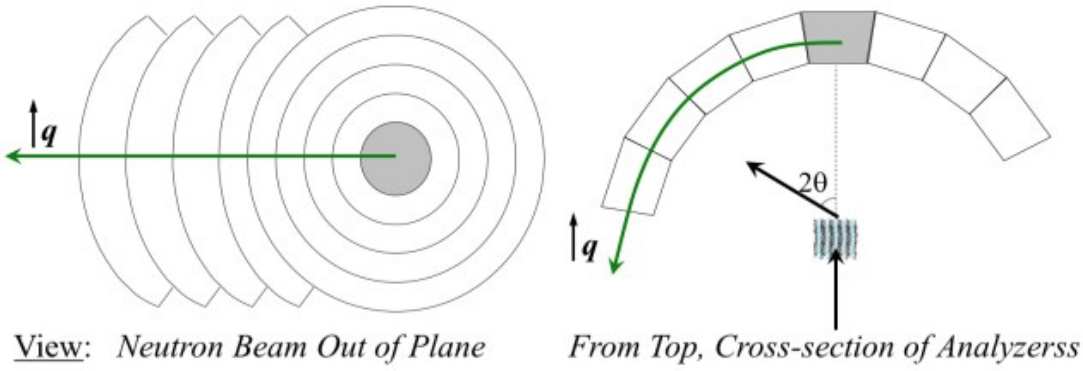
Fig. S1. Photograph of sample holder used for experimental measurements.

π

Fig. S2. Diagram showing sample orientation relative to neutron beam, arrangement of analyzers.



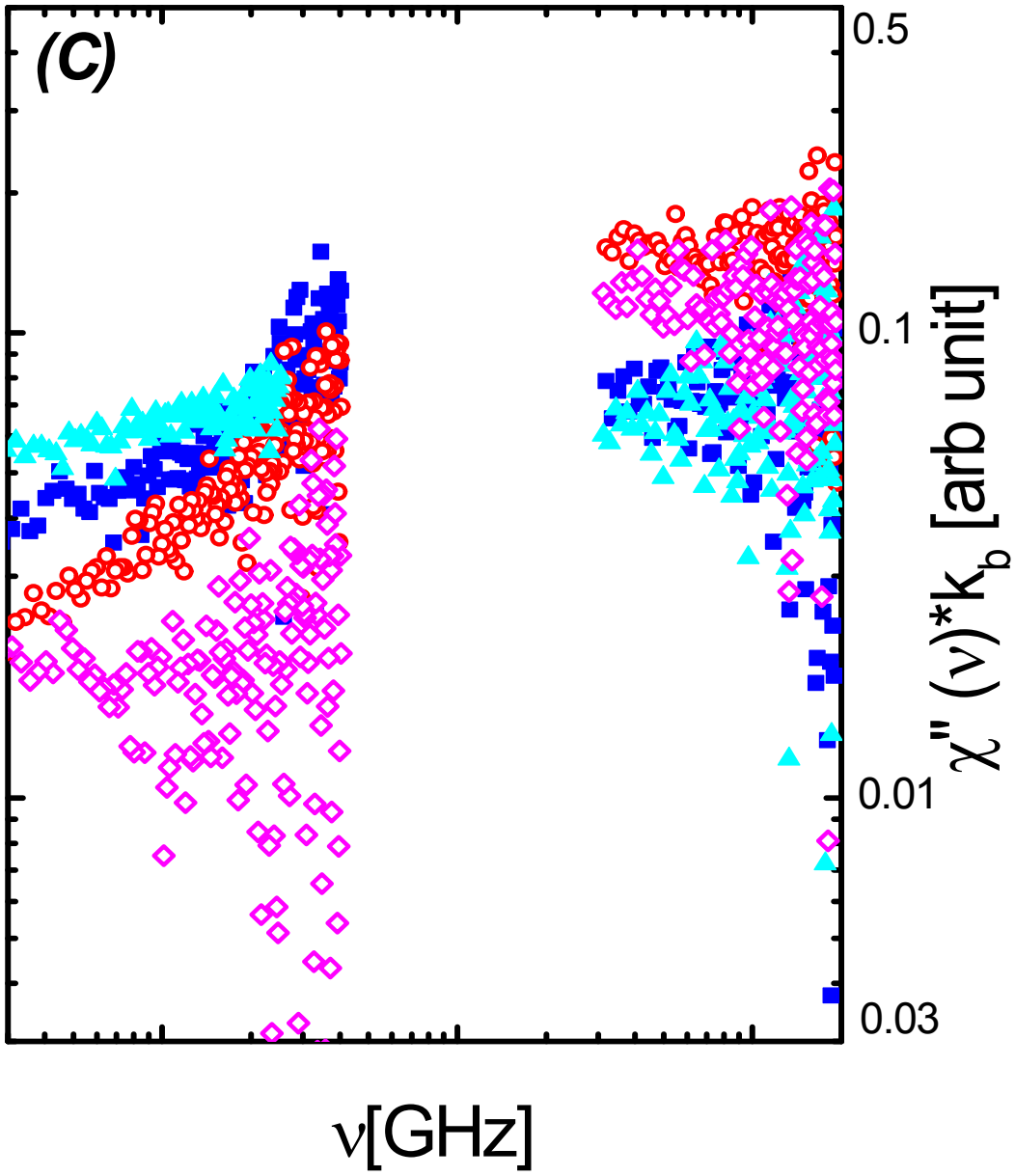


Change in momentum, *q*, is related to the distance and direction of the motion resulting in the scattered beam:

$$\vec{q} = \vec{k}_f - \vec{k}_i$$

$$q = \frac{4\pi}{\lambda}\sin(\theta)$$

Fig. S3. The imaginary part of the dynamic susceptibility, χ"(ν) of DOPC and DMPC at a momentum transfer Q=1.05 Å^{-1}.



We represent the neutron scattering data as the imaginary part of the susceptibility $\chi''(\nu) = S(Q, \nu)/n(\nu)$, where $S(Q, \nu)$ is the dynamic structure factor and $n(\nu) = [\exp(h\nu/k_bT)-1]^{-1}$ is the Bose factor. Since $S(Q,\nu)/n(\nu)$ was measured using two separate instruments, $\chi''(Q, \nu)$ data were merged using the Cole-Cole distribution function:

$$\chi''(\omega) = \chi_o \frac{(\omega\tau)^{\alpha} cos(\frac{\alpha\pi}{2})}{1 + 2(\omega\tau)^{\alpha}\sin\left(\frac{\alpha\pi}{2}\right) + (\omega\tau)^{2\alpha}} \qquad (1)$$

where $\omega=2\pi\nu$, χ_o is the amplitude and τ and α are the relaxation time and stretching parameters respectively. The relaxation times and stretching parameters were taken from fits of time-dependent data derived from $S(Q,\nu)$. It is plausible that processes may exist both outside and between the frequency ranges of the instruments used. This is manifested as a tail of a relaxation process in the frequency window of DCS ($\nu > 30$ GHz) and the predicted existence of a peak in χ'' between 4-30 GHz.

Table S1. Fitting parameter of *I(Q,t)* fitting parameters

$y = y_o + (1 - y_o)e^{(-(t/\tau)^{\beta})} : 0 \leq y_o \leq 1, 0 \leq \beta \leq 1$

Lipid	T [K]	Q [Å^{-1}]	y_0	τ [ps]	β
DOPC	245	0.77	0.63±0.00	83.88±2.99	0.46±0.01
		1.05	0.53±0.00	75.19±2.50	0.34±0.00
		1.61	0.34±0.00	30.58±1.75	0.26±0.00
DOPC	274	0.77	0.13±0.01	258.60±9.08	0.43±0.01
		1.05	0.03±0.01	142.17±4.77	0.38±0.01
		1.61	0.00±0.01	31.92±1.43	0.31±0.01
DMPC	295	0.77	0.64±0.00	73.96±3.82	0.36±0.01
		1.05	0.50±0.00	55.03±3.02	0.30±0.01
		1.61	0.34±0.00	8.25±0.21	0.26±0.00
DMPC	315	0.77	0.33±0.00	49.06±1.83	0.55±0.01
		1.05	0.27±0.00	24.15±0.50	0.40±0.01
		1.61	0.15±0.00	5.70±0.03	0.35±0.00

Figure S4. (a) Q-dependence of the average relaxation time <τ> for DOPC and DMPC and temperatures above the Tm. (b) Temperature dependence the average relaxation time <τ> for DOPC and DMPC at a representative value of Q=1.05Å^{-1}. Error bars represent one standard deviation throughout the text.

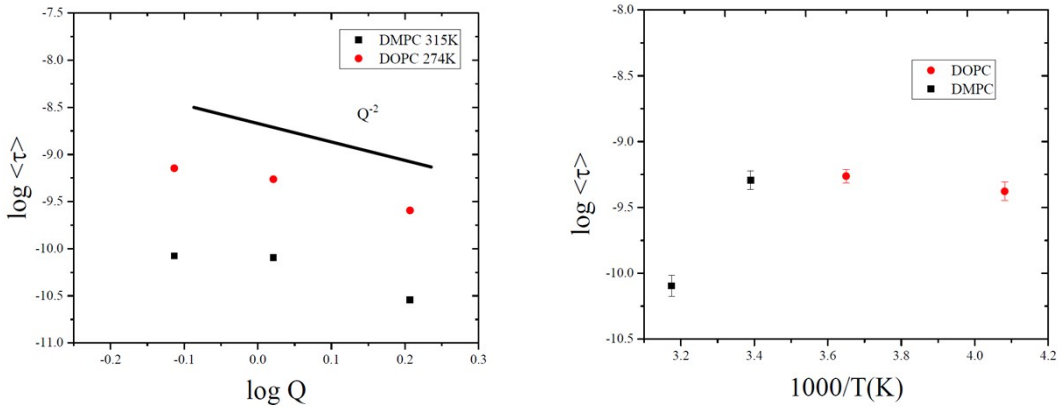


Figure S5. Q-dependence of stretching parameter β for DOPC and DMPC and temperatures above the Tm. Error bars are smaller than the data point.

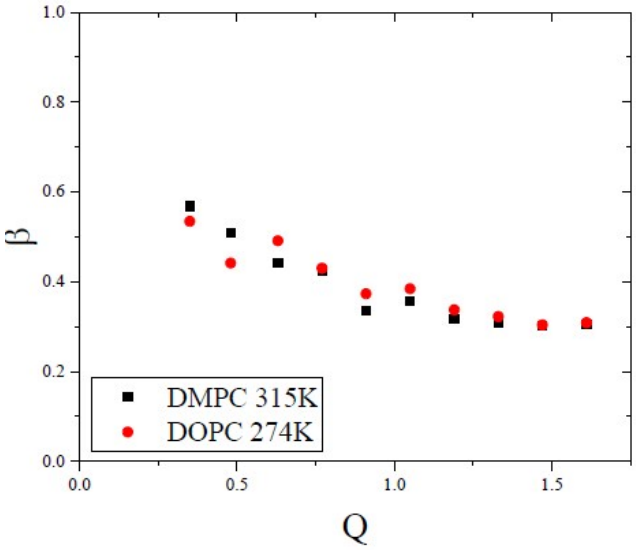


Table S2. Fitting parameters of EISF

DMPC 315K						
Q	EISF	error	1sphere fit	2 sphere fit	3 spheres	14 spheres
0.35	0.435365	0.008172	0.50779	0.44495	0.44168	0.45852
0.48	0.298048	0.008628	0.2613	0.26307	0.29325	0.25734
0.63	0.218727	0.013532	0.077292	0.14646	0.14946	0.13481
0.77	0.016372	0.007523	0.0095178	0.074278	0.05927	0.073537
0.91	0.047022	0.013127	0.00054759	0.02589	0.014343	0.038409
1.05	0	0	0.0063267	0.0048361	0.00055636	0.019196
1.19	0.0221	7.29E-03	6.65E-03	6.29E-04	0.0015921	9.01E-03
1.33	0.01712	0.005591	0.0025426	0.0018008	0.0044905	0.0041333
1.47	0.024068	9.84E-03	9.1693E-05	0.0035641	0.0046529	0.0021037
1.61	0.04934	0.00494	0.00049454	0.0035999	0.002522	0.0014272
1.76	0.055123	0.010487	0.0012621	0.0018769	0.00051231	0.0012865
DMPC 295K						
0.35	0.73688	0.006802	0.82899	0.77114		0.77487
0.48	0.647235	0.013008	0.69968	0.61584		0.58707
0.63	0.37002	0.01355	0.5338	0.43967		0.39718
0.77	0.37002	0.01355	0.38216	0.30167		0.26658
0.91	0.375784	0.015599	0.24921	0.1985		0.17854
1.05	0.289376	0.012643	0.14454	0.12743		0.12046
1.19	0.049015	2.26E-03	7.14E-02	8.00E-02		8.09E-02
1.33	0.075169	0.002342	0.02737	0.04767		0.0532
1.47	0.138578	2.80E-02	0.00623	0.02526		0.03413

1.61	0.063803	0.019303	0.00011	0.01052		0.02144
1.76	0.1327	0.018919	0.00178	0.00222		0.01276

DOPC 274K						
Q	EISF	error	1sphere fit	2 sphere fit	3 spheres	14 spheres
0.35	0.5405	0.01711	0.62214	0.52061	0.54106	0.51948
0.48	0.204	0.04485	0.39715	0.32375	0.34879	0.30909
0.63	0.1477	0.008455	0.18435	0.18528	0.19596	0.16994
0.77	0.1238	0.0075	0.062646	0.10791	0.09826	0.09893
0.91	0.05331	0.007682	0.010096	0.05253	0.03546	0.05603
1.05	0.03752	0.005678	4.78E-05	0.01734	0.00664	0.03065
1.19	0.003081	0.00537	0.004581	0.00285	0.00041	0.01609
1.33	0	0	0.007425	0.00065	0.00234	0.00801
1.47	0	0	0.005343	0.00204	0.00464	0.00395
1.61	0	0	0.001802	0.00344	0.00471	0.00213
1.76	0.004122	0.01042	3.28E-05	0.00366	0.00263	0.00143
DOPC 245						
0.35						
0.48						
0.63	0.694	0.008125	0.79402	0.72203		0.73548
0.77	0.6329	0.003019	0.70645	0.63163		0.64192
0.91	0.5227	0.004647	0.61232	0.55134		0.55438
1.05	0.5325	0.005213	0.5161	0.48579		0.47927
1.19	0.441	0.006675	0.42202	0.43577		0.42034
1.33	0.3991	0.009239	0.33374	0.39902		0.3785

1.47	0.4203	0.006813	0.25417	0.37155		0.35231
1.61	0.3461	0.008646	0.18532	0.34903		0.33867
1.76	0.261	0.007623	0.12468	0.32643		0.33361

	DMPC 315		**DMPC 295**		**DOPC 274**		**DOPC 245**	
Q	beta	error	beta	error	beta	error	beta	error
0.35	0.568233	0.011787	0.61680233	0.0238834	0.5344		0	
0.48	0.509111	0.008488	0.51997656	0.0224673	0.4415		0.3636	
0.63	0.442117	0.012313	0.40457556	0.0120175	0.4913		0.448	
0.77	0.424007	0.006202	0.40457556	0.0120175	0.4304		0.4673	
0.91	0.334873	0.007683	0.31877574	0.0115741	0.3732		0.3642	
1.05	0.35599	0.002925	0.35365638	0.0097165	0.3845		0.3554	
1.19	0.318	0.005966	0.23416267	0.0028341	0.3378		0.2896	
1.33	0.3087	0.005493	0.24050076	0.002999	0.3227		0.253	
1.47	0.302052	0.006191	0.25634995	0.0077993	0.304		0.282	
1.61	0.304693	0.006174	0.22540669	0.0049527	0.3091		0.2695	