



Figure S1. Examples of dissociation of α -synuclein when buffer solution is injected following the injection of protein. At time=0 the injection of protein is exchanged for a pure buffer solution. Data shows raw data with fits for overtone numbers 5,7,9 (top panel) and the resulting modeled thickness from a Voight-representation (bottom panels).



Figure S2. Illustration of resulting SLD distribution from selected isotope compositions for the bilayers studied with NR.



Figure S3. Thickness, shear and viscosity obtained from a fitted Voight-representation to the QCM-D data for the protein film extending out from a DOPC:DOPS 7:3 bilayer. Error bars indicate standard error equivalent to 68% confidence interval for 3 parallel measurements with individual model fitting.



Figure S4. Overlay of data in figure 7. DOPC:DOPS 7:3 bilayer (black) with adsorbed d- α -synuclein (colored).



Figure S5. Size exclusion chromatography of monomeric a-synuclein before (solid line) and after (dashed line) freeze drying. The collected volume indicated was freeze dried, redissolved in 1 ml water and then reloaded on the same column (superdex 75).



Figure S6. A sequence of measurements in NR on the same sample cell /surface. The first and last measurements on d- α -synuclein are identical albeit two protein injections with measurements in-between.



Figure S7. Overlay of data collected in the presence of 4 μ M d- α -synuclein from Figure 7 and data collected after injection of 8 μ M of d- α -synuclein. DOPC:DOPS 7:3 bilayer with 4 μ M d- α -synuclein (black) and 8 μ M d- α -synuclein (D₂O: red, H₂O: blue).

Table S1. Parameters obtained from fits to NR profiles recorded for the surface-deposition of phospholipid bilayers, i.e. the profiles presented in Figure 3. Error-weighted sum of squared residuals, χ^2 , are given for the fitted Q-range. Standard error from bootstrap analysis are given for parameters directly minimized by least square optimization.^a Manually fitted parameters^b are given error limits based on appearance of fit quality. Hydrations and thicknesses are matched to preserve the headgroup to acyl chain stoichiometry. See Material and methods for further details on the fitting procedure.

	Thickness /Å	SLD /1e-6 Å ⁻²	Solvent volume fraction /%	Roughness /Å				
SiO2	14± 0.5 ^b	3.47	28 ± 2^{b}	2				
<i>d</i> - <i>POPC</i> : <i>POPS</i> 7:3, $\chi^2 = 13.1$								
Inner and outer headgroups	5 ± 0.5^{b}	2.16	22 ± 1 ^b	1				
Inner and outer acyl chains	15± 0.5 ^b	1.99	18± 1 ^a	2				
d-POPC:CL 85:15, $\chi^2=27.9$								
Inner and outer headgroups	5 ± 0.5^{b}	1.89	26 ± 2 ^b	1				
Inner and outer acyl chains	14± 0.5 ^b	1.98	17 ± 2^{a}	2				
DOPC:DOPS 7:3, $\chi^2 = 60.0$								
Inner and outer headgroups	5 ± 0.5^{b}	2.16	32± 2 ^b	1				
Inner and outer acyl chains	15± 0.5 ^b	-1.22	25 ± 2^{a}	2				

Table S2. Parameters obtained from fits to neutron reflectivity profiles recorded for the surface-deposited phospholipid bilayers composed of d-POPC:POPS 7:3 and d-POPC:CL 85:15 in the presence of 4 μ M deuterated α -synuclein. The changed parameters compared to the bilayer fits presented in Table 1 are shown in bold. Error-weighted sum of squared residuals, χ^2 , are given for the fitted Q-range. Standard error from bootstrap analysis are given for parameters directly minimized by least square optimization.^a Manually fitted parameters^b are given error limits based on appearance of fit quality. Error limits for derived parameters^c are calculated using standard statistical methods, taking covariance into account. Hydrations and thicknesses are matched to preserve the headgroup to acyl chain stoichiometry. See Material and methods for further details on the fitting procedure.

	Thickness /Å	SLD /1e-6 Å ⁻²	Solvent fraction / % volume	Rough-ness /Å	d-α-synuclein /%V	Lip./Prot. (v/v)	Lip./Prot. (n/n)	
d-POPC: POPS 7:3 + d- α -synuclein, $\chi^2 = 16.2$								
Outer acyl-chains	14 ± 0.5^{b}	1.99	32 ± 3^{a}	2				
Outer headgroups	5 ± 0.5^{b}	3.00 ± 0.30^{a}	28 ± 3^{c}	1	12 ± 4^{c}	5.0 ± 0.4^{c}	296 ± 24^{c}	
d-POPC:CL 85:15 + d- α -synuclein, $\chi^2 = 20.6$								
Outer acyl-chains	13 ± 0.5^{b}	1.98	32 ± 2^{a}	2				
Outer headgroups	5± 0.5 ^b	2.95 ± 0.25^{a}	32 ± 2^{c}	1	14 ± 3^{c}	3.9 ± 0.3^{c}	198±15 ^c	
DOPC: DOPS 7:3 + d- α -synuclein, $\chi^2 = 50.3$								
Outer acyl chains	$14 \pm 0.5^{\text{b}}$	-1.22	37 ± 3^{a}	2				
Outer headgroups	5 ± 0.5^{b}	3.67 ± 0.35^{a}	24 ± 3^{c}	1	$23 \pm 5^{\circ}$	$2.3 \pm 0.3^{\circ}$	136± 18 ^c	