Supporting Material

1.) Calculation of neutron density profiles in stratified layers

In each medium j ($0 \leq j \leq N$) ranging from z_j to z_{j+1}, the angle-dependent intensity profile $I(\theta_i,z)$ is the absolute square of the sum of the neutron waves ψ_+ and ψ_- propagating in positive and negative z directions, respectively: $I(\theta_i, z) = \left|\psi_+(\theta_i, z) + \psi_-(\theta_i, z)\right|^2$.

In the following we use:

$$k_j^z(\theta_i) = \sqrt{\left(k_0^z(\theta_i)\right)^2 - 4\pi SLD_j} \quad \text{and} \quad k_0^z(\theta_i) = \frac{2\pi}{\lambda}\sin\theta_i ,$$

SLD_j denotes the scattering length density of the j^{th} medium. θ_i is the angle of incidence and λ the neutron wavelength. $r_{n,m}$ and $t_{n,m}$ are the Parratt amplitude reflection and transmission coefficients, respectively, for a wave traveling from medium n to medium m.

In the first (semi-infinite) medium (silicon, $j = 0$):

$$\psi_+\left(\theta_i, z \in \left[-\infty, z_j\right]\right) = \psi_0 \exp\left(ik_j^z\left(z - z_j\right)\right)$$

$$\psi_-\left(\theta_i, \in \left[-\infty, z_j\right]\right) = \psi_0 r_{j,N} \exp\left(ik_j^z\left(z_j - z\right)\right),$$

Within the slabs ($0 < j < N$) multiple reflections have to be considered:

$$\psi_+\left(\theta_i, z \in \left[z_j, z_{j+1}\right]\right) = \psi_0 t_{0,j} \exp\left(ik_j^z\left(z - z_j\right)\right) \cdot \sum_{l=0}^{\infty}\left\{r_{j,N} r_{j,0} \exp\left(2ik_j^z\left(z_{j+1} - z_j\right)\right)\right\}^l$$

$$= \psi_0 \frac{t_{0,j} \exp\left(k_j^z\left(z - z_j\right)\right)}{1 - r_{j,N} r_{j,0} \exp\left(2ik_j^z\left(z_{j+1} - z_j\right)\right)}$$

$$\psi_-\left(\theta_i, z \in [z_j, z_{j+1}]\right) = \psi_0 t_{0,j} r_{j,N} \exp\left(ik_j^z\left(z_{j+1} - z_j\right)\right) \cdot \exp\left(ik_j^z\left(z - z_j\right)\right) \cdot \sum_{l=0}^{\infty} \left\{r_{j,N} r_{j,0} \exp\left(2ik_j^z\left(z_{j+1} - z_j\right)\right)\right\}^l$$

$$= \psi_0 \frac{t_{0,j} r_{j,N} \exp\left(ik_j^z\left(z_{j+1} - z_j\right)\right) \cdot \exp\left(k_j^z\left(z - z_j\right)\right)}{1 - r_{j,N} r_{j,0} \exp\left(2ik_j^z\left(z_{j+1} - z_j\right)\right)}$$

In the last (semi-infinite) medium (D_2O, $j = N$):

$$\psi_+\left(\theta_i, z \in [z_N, \infty]\right) = \psi_0 t_{0,N} \exp\left(ik_j^z\left(z - z_j\right)\right)$$

$$\psi_-\left(\theta_i, z \in [z_N, \infty]\right) = 0$$

2.) Absorption sensitivity of reflectometry and GINF measurements

In principle, the absorption of neutrons by a target label can be determined from the decrease in neutron reflectivity. However, in addition to the severe practical obstacles imposed by diffuse scattering and the adsorption by nuclides other than the target label, there is also a fundamental limitation to the relative statistical error with which the absorption can be quantified in neutron reflectivity.

Let N_i be the number of incident neutrons during an acquisition in total reflection configuration and $P_c << 1$ the probability of each neutron to be captured by the target label in this configuration. Then the number of neutrons reflected (under ideal conditions) from the sample is

$$N_r = N_i(1 - P_c).$$

The statistical error in the measured number of reflected neutrons is

$$\Delta_r = \sqrt{N_r} \approx \sqrt{N_i} .$$

The relative statistical error corresponding to the number of captured neutrons $N_c = N_i P_c$ is

$$\delta_r = \frac{\Delta_r}{N_c} = \frac{1}{P_c \sqrt{N_i}} .$$

Now we calculate the relative statistical error corresponding to the number of γ photons counted in a GINF experiment. The probability of detecting a neutron capture event with the γ detector is denoted with P_d. Then the statistical error corresponding to the number of detected γ photons $N_\gamma = N P_c P_d$ is

$$\Delta_\gamma = \sqrt{N_\gamma} = \sqrt{N P_c P_d} ,$$

with the relative statistical error

$$\delta_\gamma = \frac{\Delta_\gamma}{N_\gamma} = \frac{1}{\sqrt{N P_c P_d}} .$$

Dividing both relative statistical errors for the same acquisition yields:

$$\frac{\delta_\gamma}{\delta_r} = \sqrt{\frac{P_c}{P_d}} .$$

As soon as P_c becomes lower than P_d, the relative error of the reflectivity exceeds that of the GINF signal, i.e., the GINF measurement will provide higher sensitivity. In the present study P_c and P_d are both at the order of 10^{-2}, despite the high label density. In future soft-matter and biology studies at the solid-liquid interface, label densities will be substantially lower, corresponding to much lower values of P_c, while at the same time there is room for significant improvement in P_d.

3.) X-ray reflectivity of silane-functionalized substrate and supported membrane in $GdCl_3$-free buffer

Table S3A: Bare silane-functionalized silicon substrate in air

	silicon	hydrocarbon	air
ρ_e (e^-/Å^{-3})	0.71	0.32	0
d (Å)	∞	19	∞
σ (Å)	4	4	-

Table S3B: Supported membrane in $GdCl_3$-free buffer

	silicon	hydrocarbon	headgroup	water
ρ_e (e^-/Å^{-3})	0.71	0.32	0.42	0.33
d (Å)	∞	29	7	∞
σ (Å)	4	2	4	-

4.) γ-detection with improved energy resolution

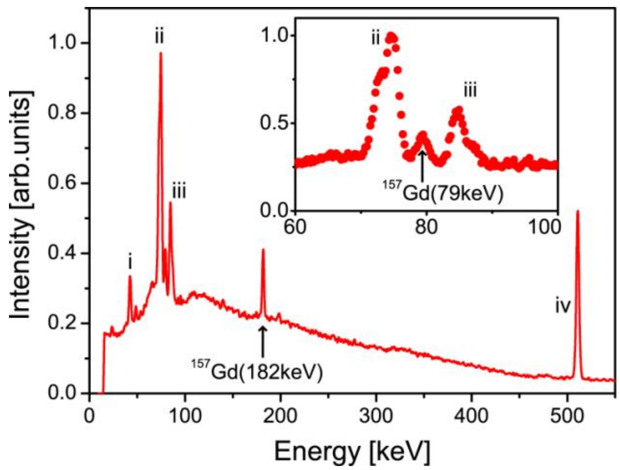

Figure S4: (main panel) γ-spectrum from a ^{157}Gd-labeled membrane with (40 mol% Gd-chelator lipid) illuminated with a neutron beam at an incident angle θ_i = 0.28 deg using γ-detection with ≈1% relative energy resolution. The characteristic fluorescence at 182 keV after neutron capture by ^{157}Gd is indicated with an arrow. Characteristic energies of internal conversion, Pb Kα, Pb Kβ, and pair annihilation are denoted i, ii, iii, and iv, respectively. The inset shows a close up view around the second strongest line of the ^{157}Gd neutron capture reaction at 79 keV. The line can be clearly identified between Pb Kα and Kβ lines.