Supplementary Material for "Nuclear Spin Alignment of Sodium Ions via Electric Field Gradients in Phospholipid Membranes"

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S1 Simulation Setup of Phospholipid Membrane

The phospholipid we used is DMPC(molecular formula: $C_{36}H_{72}NO_8P$), whose structure is shown in Fig.S 1.

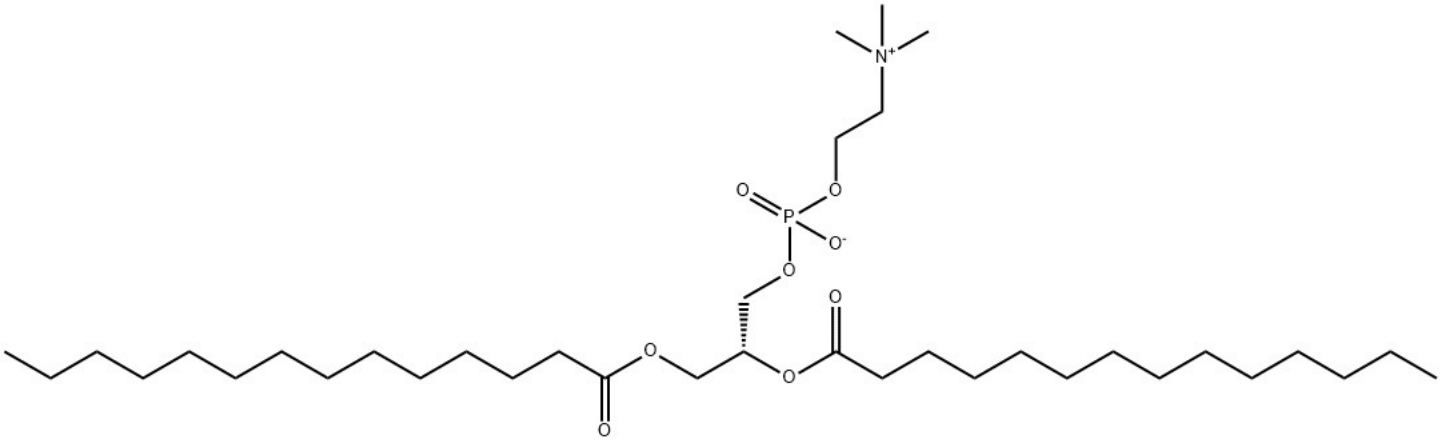

Fig. S 1. The structural formulas of DMPC.

The simulation lasted 2us and the system reached equilibrium at about 600ns, as shown in Fig.S 2. Therefore, we selected the trajectory data after 1us for our research.

S2 Rotational correlation time and diffusion coefficient

To observe the motion of the phospholipid molecules, especially the speed of rotational and diffusional motion, we examined the rotational autocorrelation curves and diffusion coefficient curves of all the phospholipid molecules. We calculated the rotational autocorrelation function of the phospholipid molecules and averaged it over the 128 molecules, resulting in the curve shown in Fig.S 3(a). Subsequently, we calculated the diffusion coefficient of the 128 phospholipid molecules (Fig.S 3(b)). The results obtained are consistent with the magnitude of some existing simulation results[1].

S3 Mathematical Calculation of the EFG

The original EFG data obtained from EFGShield program is a representation in the principal axis coordinate system:

$$V = \begin{bmatrix} V_{11} & 0 & 0 \\ 0 & V_{22} & 0 \\ 0 & 0 & V_{33} \end{bmatrix}, \tag{S1}$$

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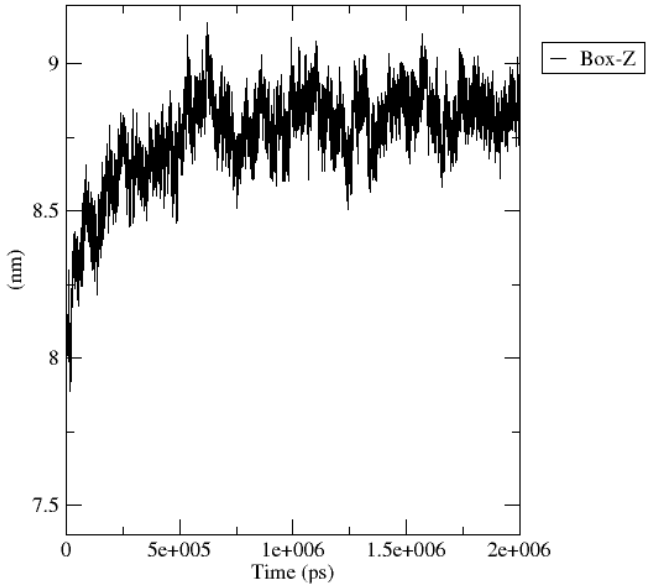
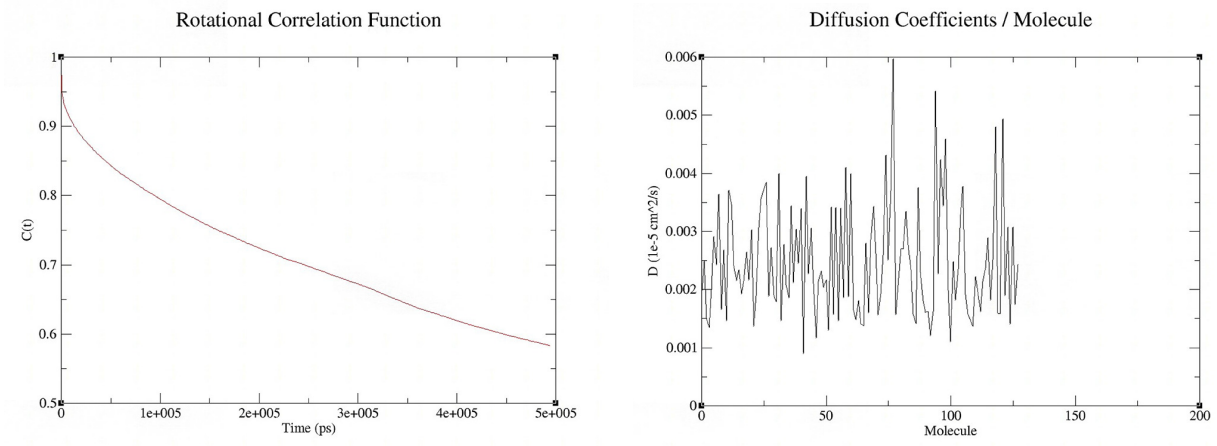


Fig. S 2. The equilibrium curve of the phospholipid membrane system with time. The thickness does not change dramatically or fluctuate after about 600ns, which implies that the system has reached equilibrium state.



(a) (b)

Fig. S 3. The rotational autocorrelation and diffusion coefficient results. (a) Average rotational autocorrelation function of 128 phospholipid molecules; (b) Diffusion coefficient of phospholipid molecules.

and three eigenvectors that represent the directions of three principal axes of the EFG respectively:

$$v_1 = \begin{pmatrix} V_{1x} & V_{1y} & V_{1z} \end{pmatrix}, \tag{S2}$$

$$v_2 = \begin{pmatrix} V_{2x} & V_{2y} & V_{2z} \end{pmatrix}, \tag{S3}$$

$$v_3 = \begin{pmatrix} V_{3x} & V_{3y} & V_{3z} \end{pmatrix}, \tag{S4}$$

while the time-averaging operation requires the EFG expressed in lipid bilayer coordinate system:

$$V^{mem} = \begin{bmatrix} V_{xx} & V_{xy} & V_{xz} \\ V_{yx} & V_{yy} & V_{yz} \\ V_{zx} & V_{zy} & V_{zz} \end{bmatrix}. \tag{S5}$$

The following relations are used to transform the two EFG:

$$V_{xx} = V_{11} \times V_{1x} \times V_{1x} + V_{22} \times V_{2x} \times V_{2x} + V_{33} \times V_{3x} \times V_{3x}, \tag{S6}$$

$$V_{xy} = V_{11} \times V_{1x} \times V_{1y} + V_{22} \times V_{2x} \times V_{2y} + V_{33} \times V_{3x} \times V_{3y}, \tag{S7}$$

$$V_{xz} = V_{11} \times V_{1x} \times V_{1z} + V_{22} \times V_{2x} \times V_{2z} + V_{33} \times V_{3x} \times V_{3z}, \tag{S8}$$

$$V_{yx} = V_{11} \times V_{1y} \times V_{1x} + V_{22} \times V_{2y} \times V_{2x} + V_{33} \times V_{3y} \times V_{3x}, \tag{S9}$$

$$V_{yy} = V_{11} \times V_{1y} \times V_{1y} + V_{22} \times V_{2y} \times V_{2y} + V_{33} \times V_{3y} \times V_{3y}, \tag{S10}$$

$$V_{yz} = V_{11} \times V_{1y} \times V_{1z} + V_{22} \times V_{2y} \times V_{2z} + V_{33} \times V_{3y} \times V_{3z}, \tag{S11}$$

$$V_{zx} = V_{11} \times V_{1z} \times V_{1x} + V_{22} \times V_{2z} \times V_{2x} + V_{33} \times V_{3z} \times V_{3x}, \tag{S12}$$

$$V_{zy} = V_{11} \times V_{1z} \times V_{1y} + V_{22} \times V_{2z} \times V_{2y} + V_{33} \times V_{3z} \times V_{3y}, \tag{S13}$$

$$V_{zz} = V_{11} \times V_{1z} \times V_{1z} + V_{22} \times V_{2z} \times V_{2z} + V_{33} \times V_{3z} \times V_{3z}. \tag{S14}$$

In the following, we use a specific example to illustrate our handling of EFG data. Suppose we obtain an EFG in atomic units as follows from EFGShield program:

$$V = \begin{bmatrix} -0.051896 & 0 & 0 \\ 0 & -0.118082 & 0 \\ 0 & 0 & 0.169978 \end{bmatrix}, \tag{S15}$$

and three eigenvectors:

$$v_1 = \begin{pmatrix} 0.927900 & 0.368271 & -0.058123 \end{pmatrix}, \tag{S16}$$

$$v_2 = \begin{pmatrix} -0.307168 & 0.843490 & 0.440650 \end{pmatrix}, \tag{S17}$$

$$v_3 = \begin{pmatrix} 0.211304 & -0.391026 & 0.895795 \end{pmatrix}. \tag{S18}$$

Transform it into the expression in lipid bilayer coordinate system according to Eq. (S6) - Eq. (S14):

$$V^{mem} = \begin{bmatrix} -0.04823 & -0.00118 & 0.050956 \\ -0.00118 & -0.06506 & -0.10232 \\ 0.05956 & -0.10232 & 0.113295 \end{bmatrix}, \tag{S19}$$

which implies the value of the EFG at the position of this sodium ion at this time. Averaging it with data at other 199 moments, an time-averaged EFG at the position of this sodium ion is obtained:

$$\overline{V}^{mem} = \begin{bmatrix} -0.04344 & -0.0013 & -0.00464 \\ -0.0013 & -0.02287 & -0.0329 \\ -0.00464 & -0.0329 & 0.066308 \end{bmatrix}. \tag{S20}$$

Finally, to obtain the directions of three principal axes of the time-averaged EFG, transform Eq. (S20) into the expression in principal axis coordinate system by diagonalizing the matrix:

$$\overline{V} = \begin{bmatrix} -0.03301 & 0 & 0 \\ 0 & -0.04426 & 0 \\ 0 & 0 & 0.077266 \end{bmatrix}, \tag{S21}$$

$$\bar{v_1} = \begin{pmatrix} -0.24599 & 0.923474 & 0.294421 \end{pmatrix}, \tag{S22}$$

$$\bar{v_2} = \begin{pmatrix} 0.968704 & 0.223836 & 0.107288 \end{pmatrix}, \tag{S23}$$

$$\bar{v_3} = \begin{pmatrix} -0.03318 & -0.3116 & 0.949635 \end{pmatrix}, \tag{S24}$$

which implies that the principal axis corresponding to $\overline{V}_{33}$ is close to z-axis. This final data form is used for our data presentation in the figures.

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