Supporting Information: NMRlipids IV: Headgroup & glycerol backbone structures, and cation binding in bilayers with PE and PG lipids

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S1 Simulated systems

S1.1 CHARMM36

POPE Simulation details by M. Javanainen.

POPE with additional NaCl Simulation details by A. Peon.

POPG Simulation details by Ollila.

POPG with additional NaCl Simulation details by A. Peon.

POPC:POPE mixtures Data is available at.[1,2] 300 K with v-rescale (tau=0.1 ps), 1 bar with PR semiisotropic (tau=4 ps, compressibility=4.5e-5 bar^{-1}), PME order 4 and space 0.12, rcoulomb and rvdw 1.0, 128 lipids per leaflet, no ion Full simulation details by Fuchs et al.

POPC:POPG mixture with additional calcium Simulation details by J. Madsen.

POPC:POPG mixture with additional NaCl Simulation details by A. Peon.

S1.2 CHARMM36ua

POPE Data is available at.[3] Simulation details by T. Piggot.

S1.3 Slipids

POPE Data is available at.[4] Simulation details by T. Piggot.

POPE with additional NaCl Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

DPPE Data is available at.[5] Simulation details by F. Favela.

POPG Data is available at.[6] Simulation details by F. Favela. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

POPG with additional NaCl Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

DPPG Data in 298 K is available at[7] and in 314 K at.[8] Simulation details by F. Favela. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not

true.

POPC:POPG mixture with additional NaCl Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

S1.4 Berger

POPE Data is available at.[9,10] Simulation details by T. Piggot.

DOPE Data is available at.[11,12] Simulation details by T. Piggot.

POPC:POPE, POPC:DOPE and DOPC:DOPE mixtures Data is available at.[13,14] 300 K with v-rescale (tau=0.1 ps), 1 bar with PR semiisotropic (tau=4 ps, compressibility=4.5e-5 bar^{-1}), PME order 4 and space 0.12, rcoulomb and rvdw 1.0, 128 lipids per leaflet, no ion Simulation details by Fuchs et al.

S1.5 GROMOS 43A1-S3

POPE Data is available at.[15] Simulation details by T. Piggot.

S1.6 OPLS-UA

POPE Data is available at.[16] Simulation details by T. Piggot.

POPE with vdW interaction in H Data is available at.[17] Simulation details by T. Piggot.

S1.7 GROMOS-CKP and GROMOS-CKPM

POPE Data is available at.[18] Simulation details by T. Piggot.

DOPE Data is available at.[19] Simulation details by T. Piggot.

DPPE Data is available at.[20] Simulation details by T. Piggot.

S1.8 Lipid17

S1.9 ECC-LIPID POPG

In ECC-lipid models, electronic continuum correction (ECC) is applied to implicitly include the missing electronic polarizability into the force field description.[21,22] In practise, this is implemented by scaling the charges and Lennard-Jones σs of headgroup, glycerol backbone, and carbonyl regions of Amber Lipid14/17 models are scaled by constant factors. Here, we follow the approach that previously improved ion binding to bilayers containing negatively charged PS lipids:[22] ECC-POPC parameters (scaling factors f_q=0.8 and f_σ=0.89 applied to Lipid14 POPC parameters)[21] were used for POPC and scaling factors of f_q=0.75 and f_σ=0.89 were applied to the charges and Lennard-Jones σs of headgroup, glycerol backbone, and carbonyl regions of Amber Lipid17 POPG parameters. The Lipid17 parameters (described above) were taken from Ref.[23] with the correct dihedral type, and the resulting parameters are available from Ref.[?] ECC-ion parameters with the scaled charges[24–26] were used in these simulations.

S2 R-PDLF and SDROSS experiments

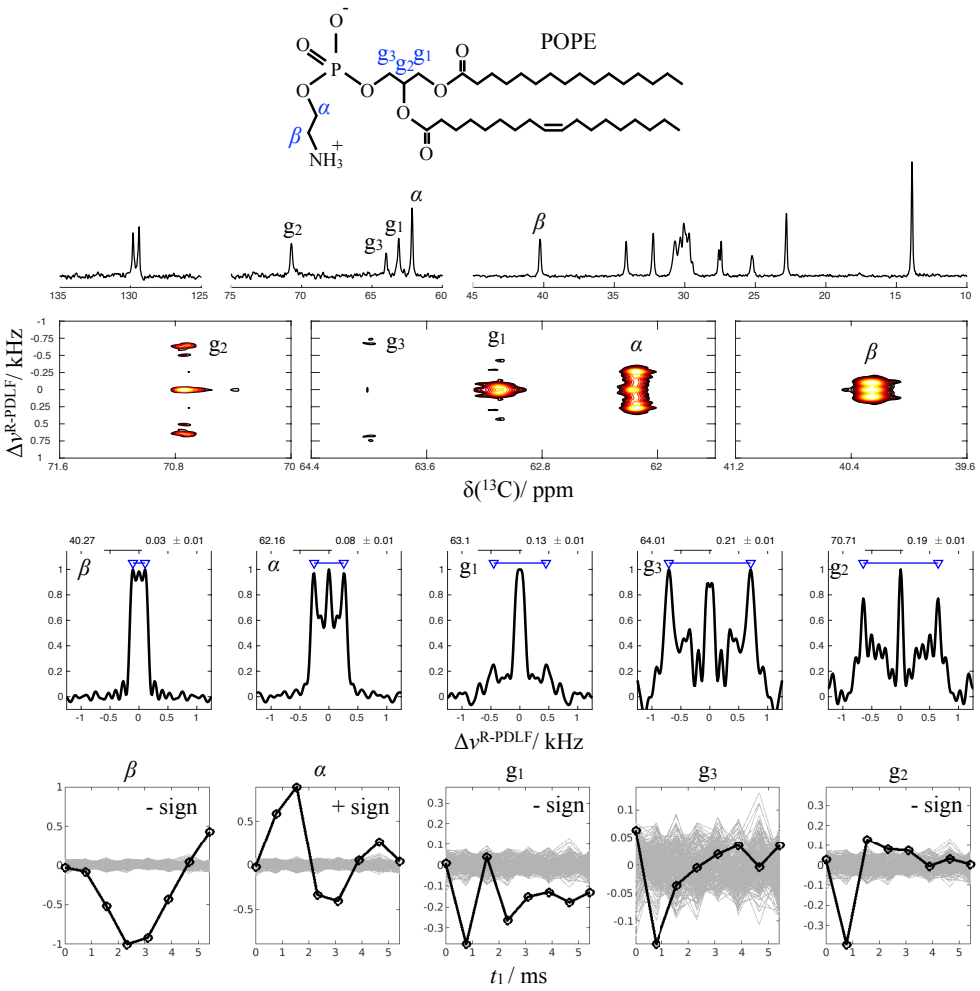

Figure S1: (A) Chemical structure of POPE with the labeling of headgroup and glycerol backbone carbons. (B) INEPT spectra from POPE sample with the headgroup and glycerol backbone peaks labeled. (C) 2D R-PDLF spectra (D) Dipolar sliced from the 2D R-PDLF spectra with the resulting order parameters on top of figures. (E) Experimetal S-DROSS curves giving signs of the order parameters.

A, B etc. labels to be put in the figure.

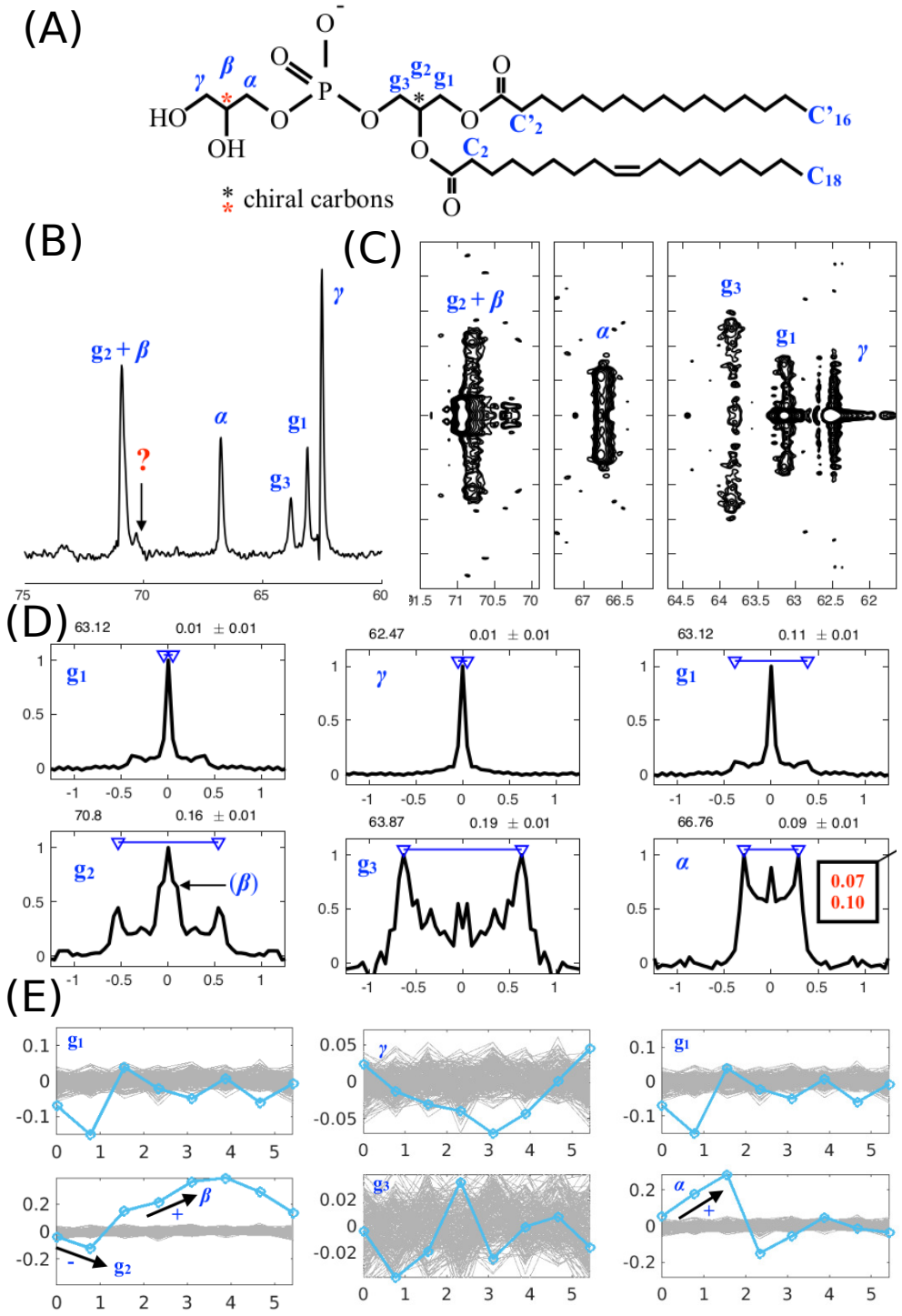

Figure S2: (A) Chemical structure of POPG with the labeling of headgroup and glycerol backbone carbons. (B) INEPT spectra from POPG sample with the headgroup and glycerol backbone peaks labeled. (C) 2D R-PDLF spectra (D) Dipolar sliced from the 2D R-PDLF spectra with the resulting order parameters on top of figures. (E) Experimetal S-DROSS curves giving signs of the order parameters.

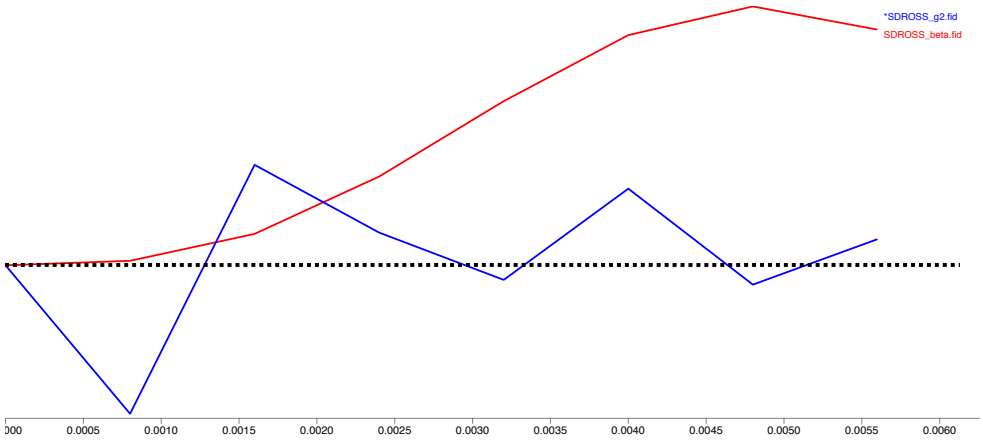

Figure S3: Simpson simulaton of S-DROSS curve of β-carbon of POPG.

S3 Changes of PG headroup order parameters upon addition of PC

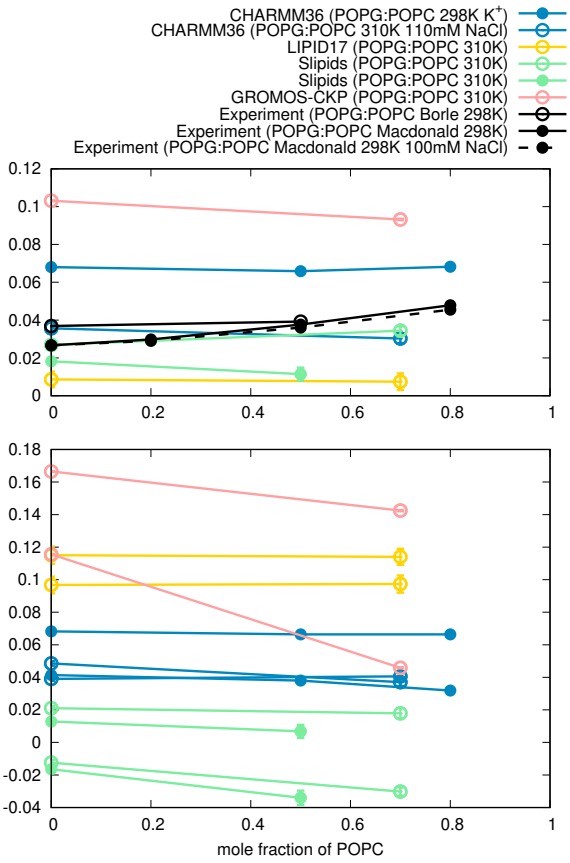

Figure S4: Modulation of PG lipid headgroup order parameters with the increasing amount of PC in lipid bilayer from experiments[27,28] and simulations with different force fields.

S4 Sodium binding to POPC simulations

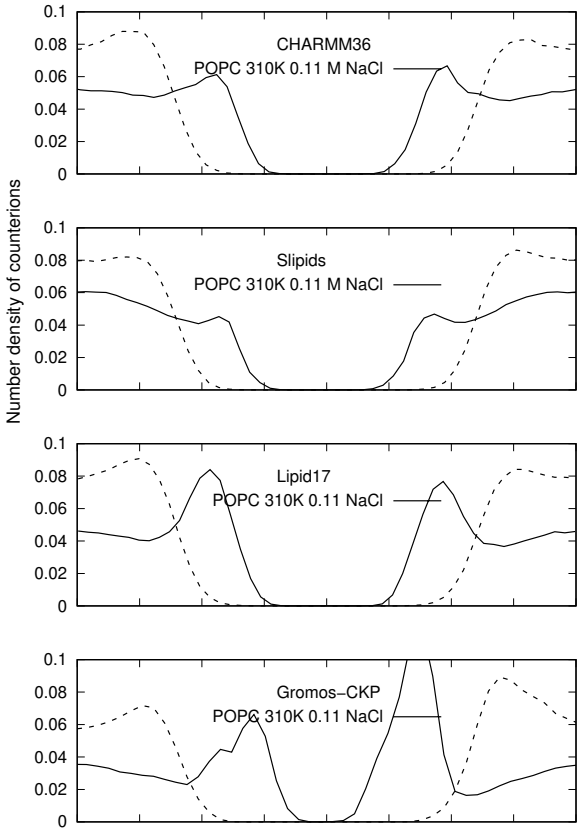

Figure S5: Sodium (solid line) and choride ion density profiles along membrane normal from different simulations with PC lipids.

Discussion about differences to the NMRlipids II to be discussed once we have the details on ions models.

S5 Calcium binding to POPC:POPG (4:1) mixtures

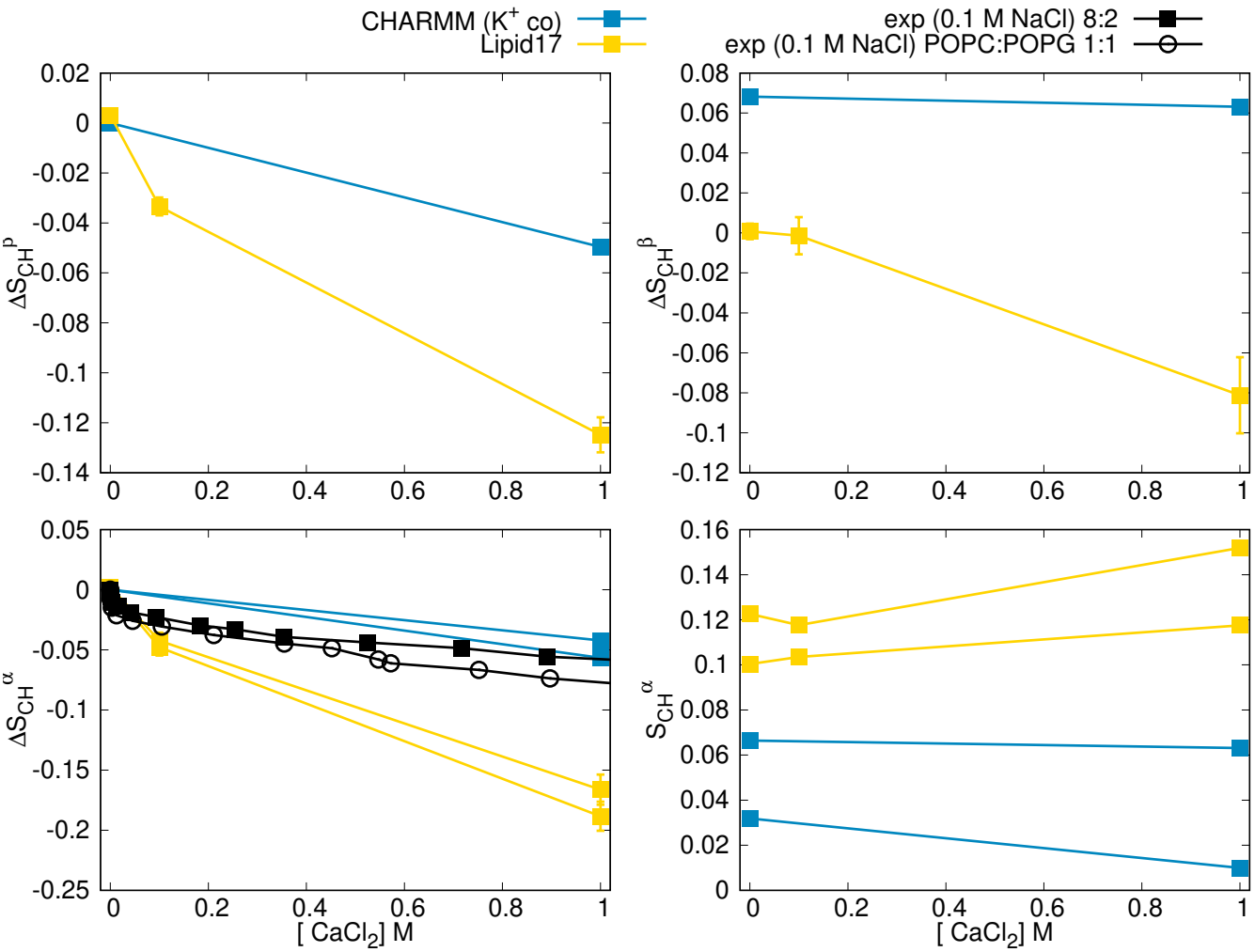

Figure S6: Modulation of headgroup order parameters of POPC (*left*) and POPG (*right*) in POPC:POPG (4:1) mixture upon addition of $CaCl_2$ in 298 K temperature from experiments[28] and simulations. The changes with respect to the systems without $CaCl_2$ are shown for other data than for the α-carbon of POPG for which experimental order parameter is not available. CHARMM36 simulations should be longer and with Na couterions.

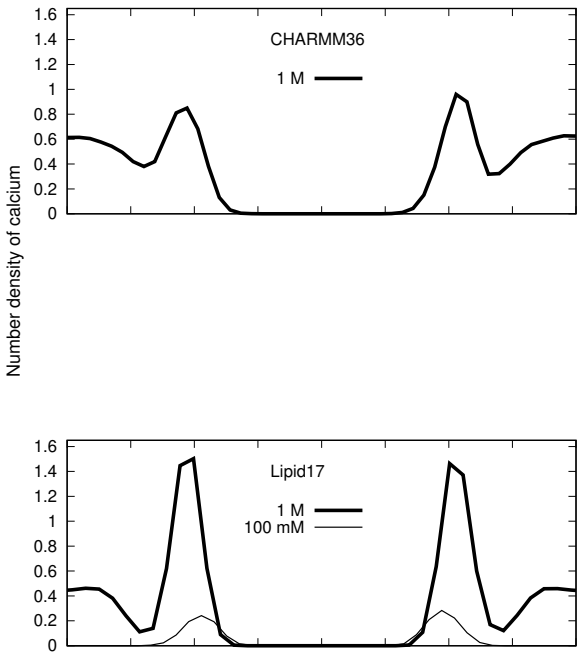

Figure S7: Calcium ion density profiles along membrane normal from simulations of POPC:POPG (4:1) mixtures with different force fields.

References

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