Supporting Information for Influence of single-stranded DNA Coatings on the Interaction Between Graphene Nanoflakes and Lipid Bilayers

Timothy C. Moore,[1,2†] Alexander H. Yang,[1,2] Olu Ogungbesan,[1,2,3] Remco Hartkamp,[1,2,††] Christopher R. Iacovella,[1,2] Qi Zhang,[4†††] and Clare McCabe[,1,2,5]*

[1]Department of Chemical and Biomolecular Engineering, Vanderbilt University, Nashville, Tennessee, USA

[2]Multiscale Modeling and Simulation (MuMS) Center, Vanderbilt University, Nashville, Tennessee, USA

[3]Department of Chemical, Biochemical, and Environmental Engineering, University of Maryland Baltimore County, Baltimore, Maryland, USA

[4]Department of Pharmacology, Vanderbilt University, Nashville, Tennessee, USA

[5]Department of Chemistry, Vanderbilt University, Nashville, Tennessee, USA

Corresponding Author

Clare McCabe, c.mcabe@vanderbilt.edu, 615-322-6853

Present Addresses

†T.C. Moore, Department of Chemical Engineering, University of Michigan, Ann Arbor, Michigan, USA

†† R. Hartkamp, Department of Process and Engineering, TU Delft, Delft, Netherlands

††† Q. Zhang, Brain Institute, Florida Atlantic University, Jupiter, Florida, USA

INITIALIZATION OF GNFS WITH DNA COATING

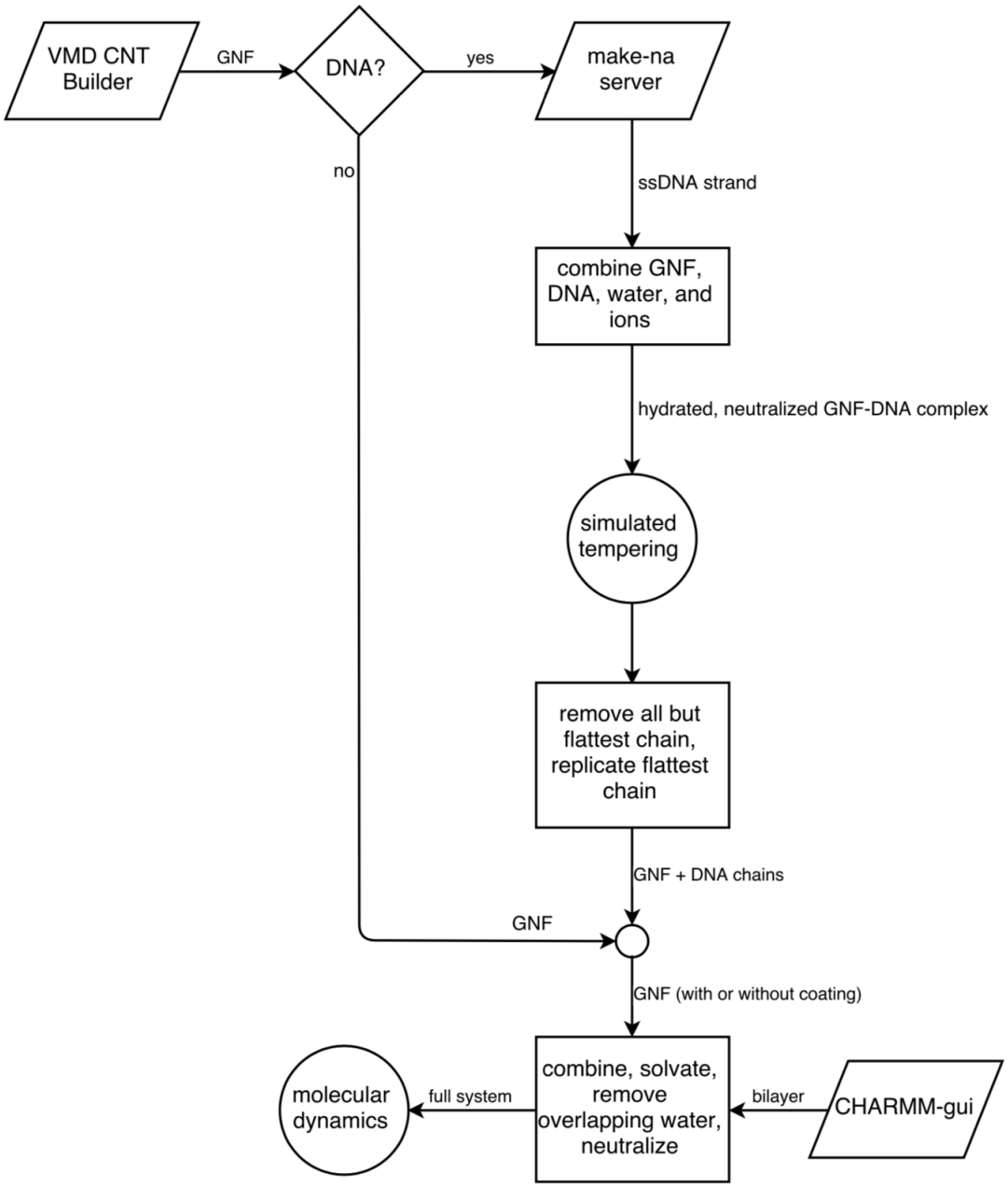

Figure S1: Process flow for initializing and simulating the systems studied in this work. Parallelograms represent external programs used to generate structures, diamonds represent decision points, rectangles represent structure manipulation, and large circles represent simulations.

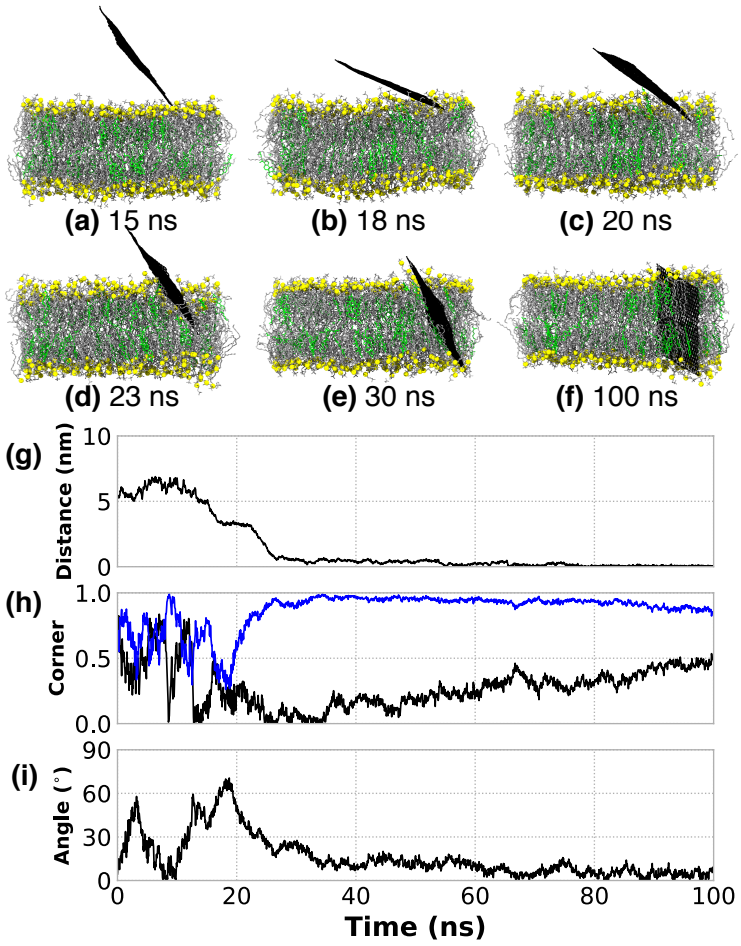

Figure S2: Insertion of a bare GNF where the GNF initially approaches the bilayer at a non-ideal angle of insertion. (a-f) snapshots of the insertion process as a function of time. (g) The distance between the centers-of-mass of the GNF and bilayer, projected along the bilayer normal; the plateau in separation between ~17-27 ns corresponds to the reorientation of the GNF before insertion into the bilayer. (h) The normalized dot product calculated between the vectors describing the GNF diagonals (blue and black) and the bilayer normal, demonstrating the corner first insertion at ~27 ns. (i) The angle between the GNF and bilayer normal is plotted as function of time, showing the reorientation of the GNF during insertion, where the value slowly achieves a value close to 0° at the end of the simulation.

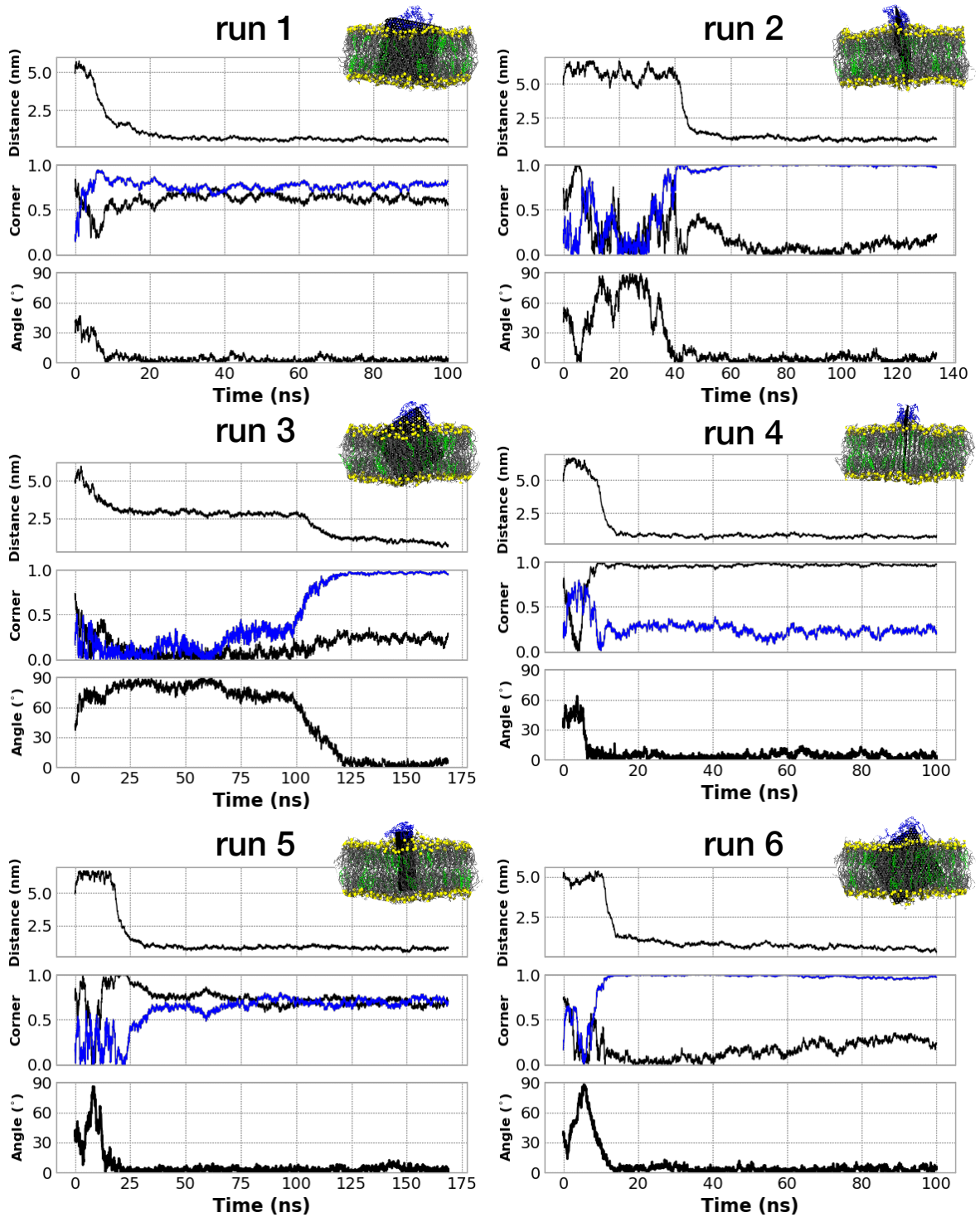

Figure S3: Analysis of 6 replicates for the 2 ssDNA coated GNF simulations. For each replicate, the following metrics are calculated: the distance between the centers-of-mass of the GNF and

bilayer projected along the bilayer normal ("Distance"); the normalized dot product calculated between the vectors describing the GNF diagonals (blue and black) and the bilayer normal, where a value of unity for one of the diagonals represents a corner first orientation ("Corner"); and angle between the GNF and bilayer normal where a value of 0° corresponds to the perpendicular orientation of the GNF sheet and a value of 90° to the GNF lying flat upon the bilayer interface ("Sheet"). A simulation snapshot corresponding to the final configuration of the simulation is inset for each replicate, where water is not shown for clarity, P atoms of DOPC are shown as yellow to highlight boundary, DOPC tails are shown as gray, and CHOL colored green, following the color scheme in the main text.

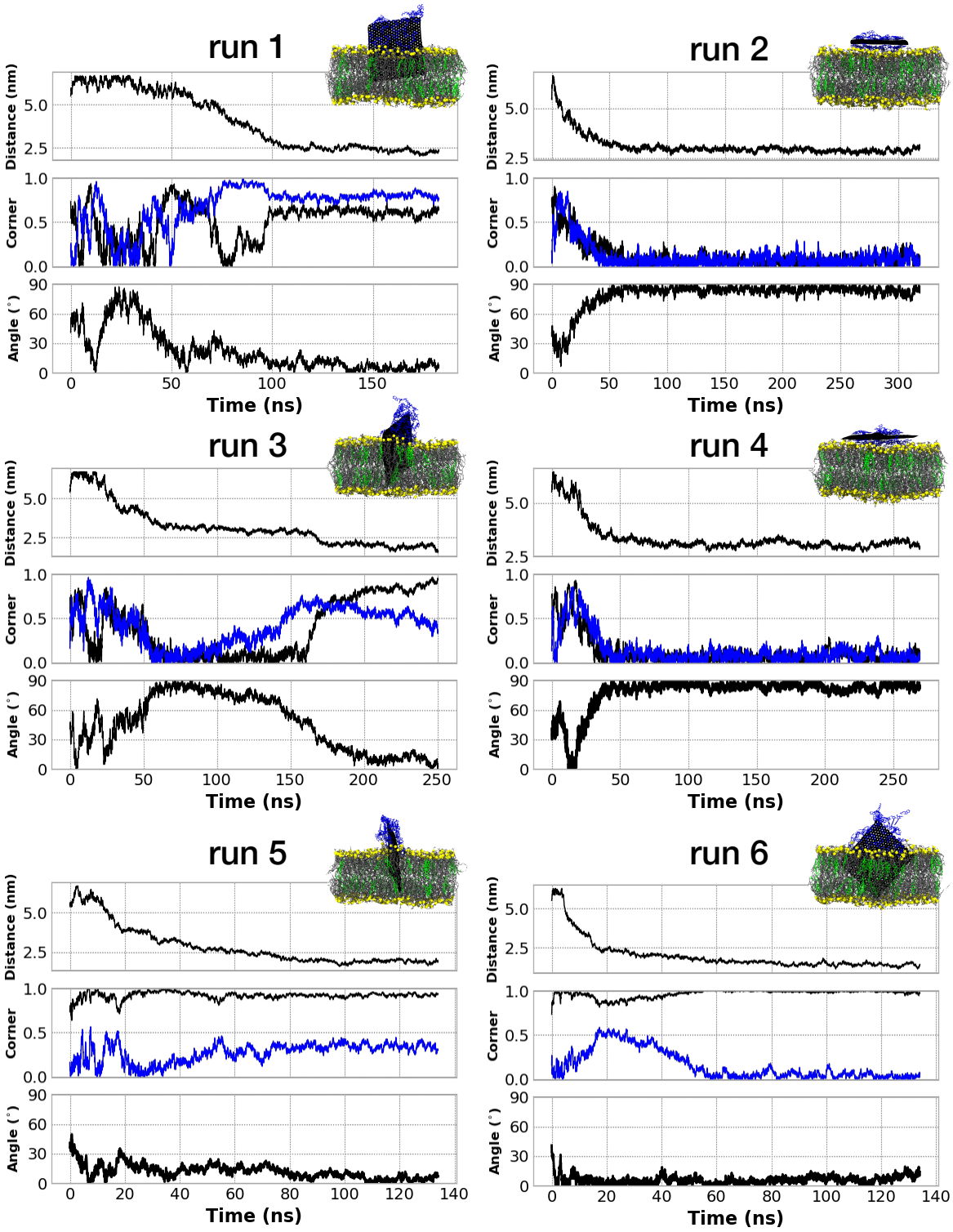

Figure S4: Analysis of 6 replicates for the 4 ssDNA coated GNF simulations. The analysis and color scheme follows Fig. S3.

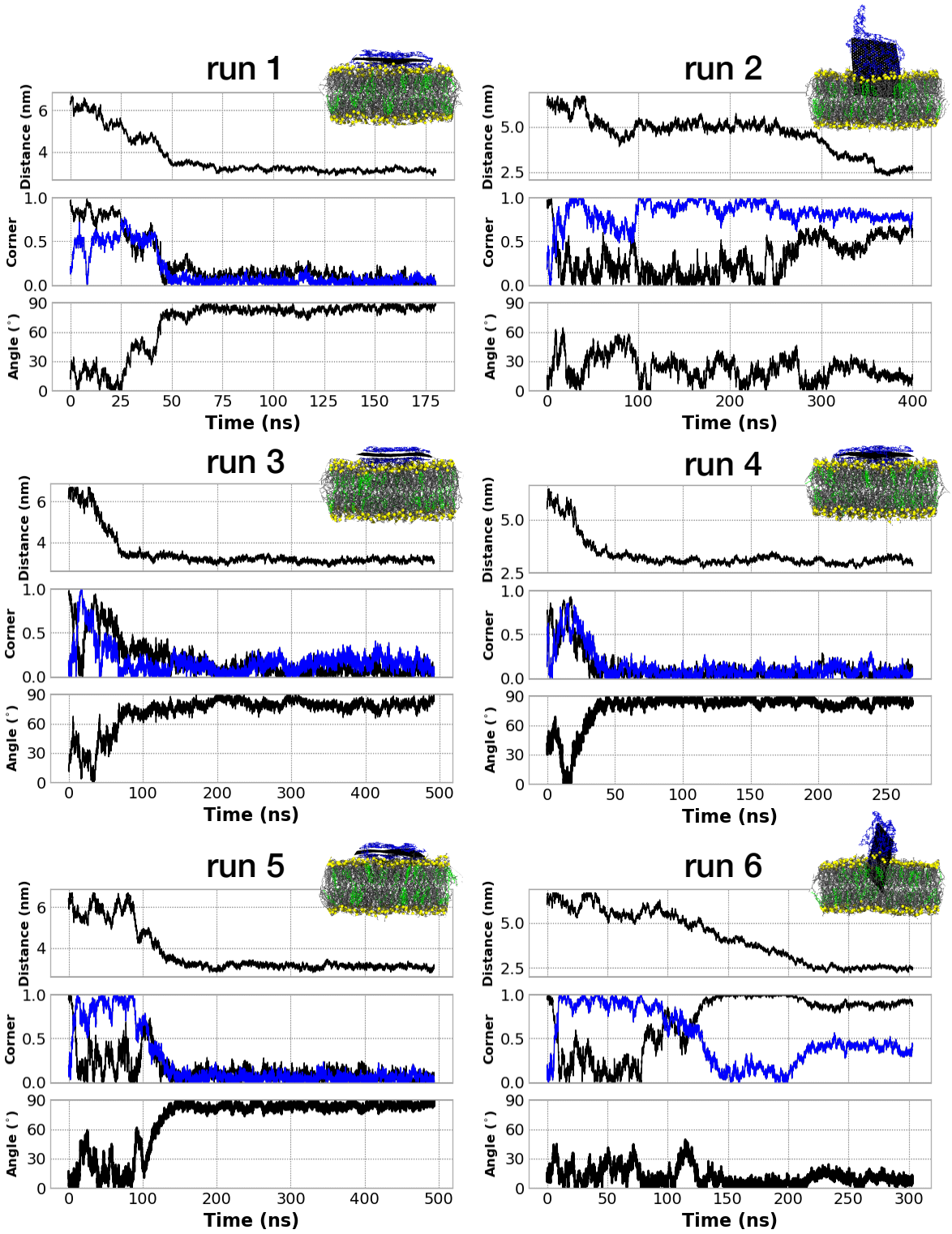

Figure S5: Analysis of 6 replicates for the 6 ssDNA coated GNF simulations. The analysis and color scheme follows Fig. S3.

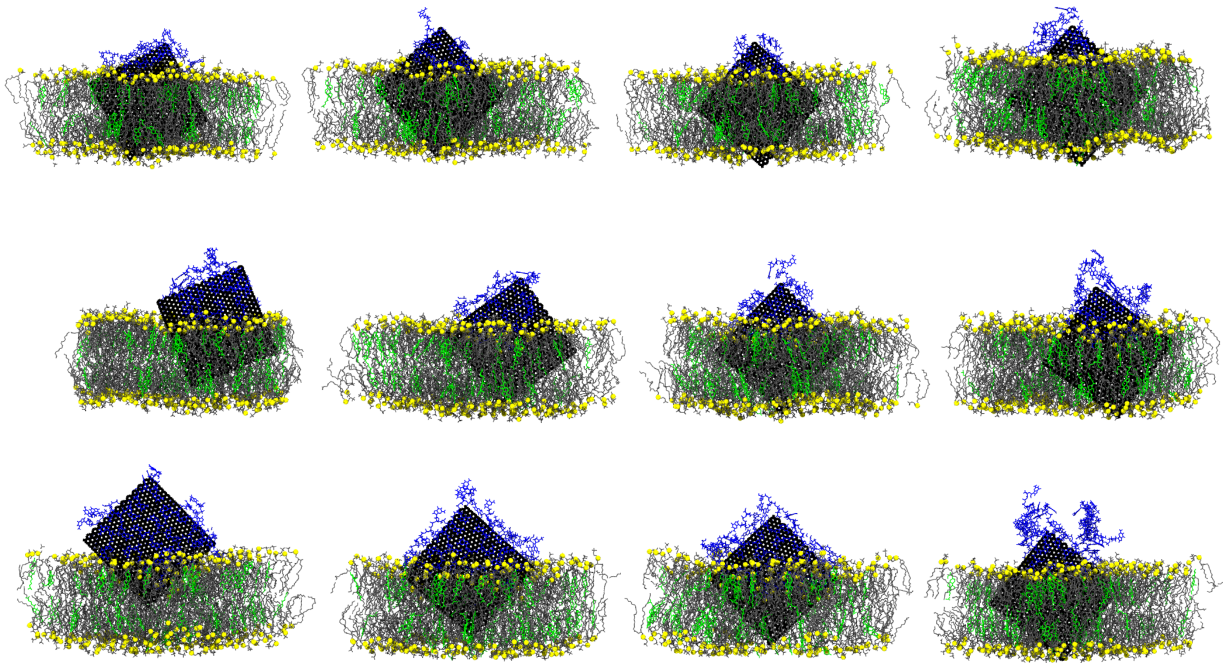

Figure S6. Final configurations of steered simulations. From left to right: various spring constants of 50, 125, 250, and 500 kJ mol-1 nm-2. From top to bottom: coatings of 2, 4 and 6 ssDNA.

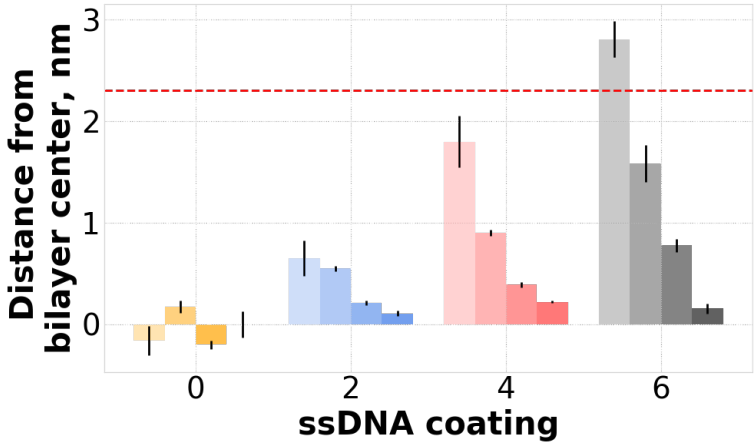

Figure S7. Distance between the centers-of-mass of the GNF and bilayer at the end of the steered simulation four different ssDNA coatings. Four different spring constants, 50, 125, 250, and 500 kJ $mol^{-1} nm^{-2}$ are presented respectively (darker shades are stiffer constants). Bilayer half-thickness is shown as a red dashed line for reference. Color code: 0 ssDNA, i.e. bare GNF (orange), 2 ssDNA (blue), 4 ssDNA (red), 6 ssDNA (black).