Supplementary Information (SI) for Soft Matter.

Electronic Supplementary Information for *Soft Matter* manuscript: Shear annealing of self-interacting sheets

William T. Funkenbusch[a], Kevin S. Silmore[a], and Patrick S. Doyle[a]

[a]*Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge MA, 02139 (USA)*

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SI.01: Sample videos of different protocols

Sheets are hexagonal with circumradius $L = 39a$, where a is the radius of the constitute beads. The beads are in a triangular grid such that each bead has 6 tangent neighbors, totaling $N = 1141$ beads. Sheets were initially in a flat configuration such that all beads lie in the flow-vorticity plane, then rotated by an angle $\theta = 5°$ about the vorticity axis and a variable angle ϕ about the flow axis. Non-neighboring beads were given a hard-sphere potential, while neighbors were given a harmonic potential with spring constant k. Neighboring triangles of three beads (such that they share two beads) were given a dihedral potential with bending rigidity κ. Finally, short-range interactions were applied in the form of a truncated Lennard-Jones potential with interaction strength ε and interaction range $\sigma = 4\sqrt{6}a/3$. All simulations were run with a time step of $\dot{\gamma}\Delta t = 2 \times 10^{-4}$.

The system was integrated in time using an Euler-Maruyama integrator for Brownian dynamics with hydrodynamics. Please see the main article and previous work[1] for details on the potentials, integration scheme, and software.

The phase space is defined by three parameters: χ^{-1}, K, and ϕ. χ^{-1} measures the relative strength of shear to interactions and is given by

$$\chi^{-1} = \frac{6\pi\eta\dot{\gamma}L^2\sigma}{\tilde{\varepsilon}\sigma^2}\left(\frac{L}{a}\right)^{-1}$$

where η is the viscosity, $\tilde{\varepsilon} = \varepsilon/2\sqrt{3}a^2$ is the interaction energy density, and all other parameters are as described earlier. Please see Appendix A of the main article for a derivation of this dimensionless group. K measures the relative strength of bending rigidity to interaction strength and is given by

$$K = \frac{\kappa}{\tilde{\varepsilon}\sigma^2}$$

where κ is the bending rigidity, as described earlier. Finally, ϕ is the initial orientation of the sheet relative to the flow axis, as described earlier.

On the left side of each video is the sheet simulation showing every $1\dot{\gamma}t$. All simulation videos were rendered in Ovito[2]. The x-axis is the flow direction, the y-axis is the shear direction, and the z-axis is the vorticity axis. On the right side of each video is the corresponding signed local mean curvature of the sheet. Please see the main article for details on this quantity.

In linear annealing simulations, the simulation is run at an initial $\chi^{-1} = 4.1 \times 10^2$ and various K and ϕ for $2000\dot{\gamma}_0 t$, where $\dot{\gamma}_0$ is the initial shear rate. Then, the shear rate is lowered linearly over a variable t_q at constant K. Finally, a no shear simulation is run for another $2000\dot{\gamma}_0 t$, again at constant K.

In Protocol 1 simulations, the simulation is run at an initial $\chi^{-1} = 4.1 \times 10^1$ and various K and ϕ for $2000\dot{\gamma} t$. Then, χ^{-1} is set to 1.4×10^2 at constant K for another $2000\dot{\gamma} t$. Finally, χ^{-1} is set to 1.4×10^1 at constant K for another $2000\dot{\gamma} t$.

In Protocol 2 simulations, the simulation is run at an initial $\chi^{-1} = 4.1 \times 10^3$ and various K and ϕ for $2000\dot{\gamma} t$. Then, χ^{-1} is set to 1.4×10^2 at constant K for another $2000\dot{\gamma} t$. Finally, χ^{-1} is set to 1.4×10^1 at constant K for another $2000\dot{\gamma} t$.

annealing_full.mp4 – Example linear annealing simulation with $K = 0.03$, $\phi = 0°$, and $t_q = 1000\dot{\gamma}_0 t$.

protocol_1_full.mp4 – Example Protocol 1 simulation with $K = 0.03$ and $\phi = 0°$.

protocol_2_full.mp4 – Example Protocol 2 simulation with $K = 0.03$ and $\phi = 5°$

high_alignment_teacup_full.mp4 – Example Protocol 2 simulation with $K = 0.03$ and $\phi = 0°$ which results in a high alignment, 2D folded sheet due to teacup behavior as described in previous work[1].

SI.02: Summary statistics for all Protocol 1 simulations

The radius of gyration is given by the square root of the sum of the eigenvalues of the gyration tensor:

$$R_g = \sqrt{\sum \lambda_i^2}$$

The radius of gyration is a measure of the overall **size** of a sheet. The relative shape anisotropy can similarly be defined in terms of the eigenvalues of the gyration tensor:

$$\zeta^2 = \frac{3}{2}\frac{\sum \lambda_i^4}{(\sum \lambda_i^2)^2} - \frac{1}{2}$$

The relative shape anisotropy is bounded between 0 and 1. $\zeta^2 = 0$ only if the beads are spherically symmetric and $\zeta^2 = 1$ only if the beads are all colinear. The relative shape anisotropy is therefore a measure of the **shape** of a sheet.

The alignment is given by the magnitude of the dot product of the eigenvector corresponding to the largest eigenvalue of the gyration tensor, $\boldsymbol{v}_1$, with the vorticity axis, $\hat{z}$: $|\boldsymbol{v}_1 \cdot \hat{z}|$. The alignment is therefore a measure of the **orientation** of a sheet.

In the main article, we show only averages over all initial conditions. Here, we give results for all initial conditions, grouped by protocol and the value of K.

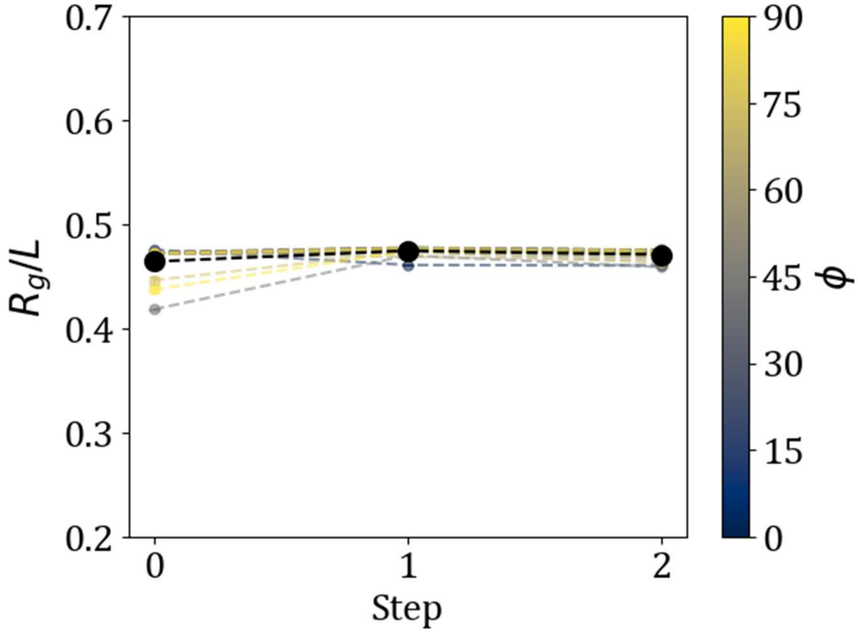

Figure S02.01: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

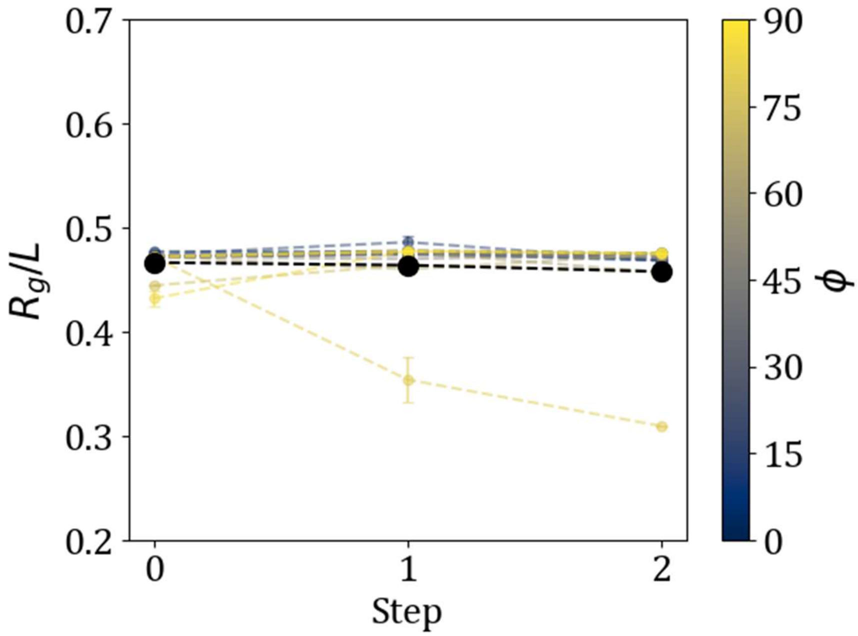

Figure S02.02: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

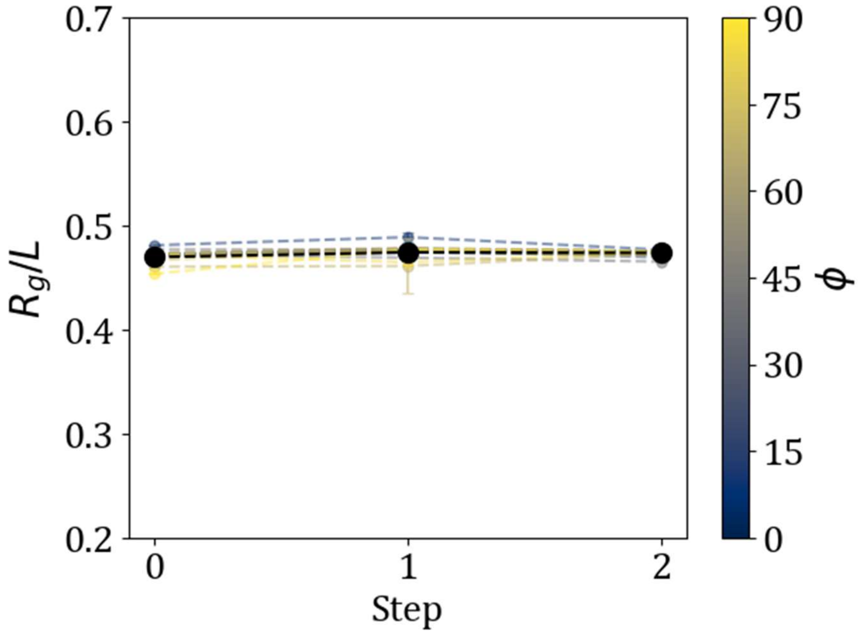

Figure S02.03: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

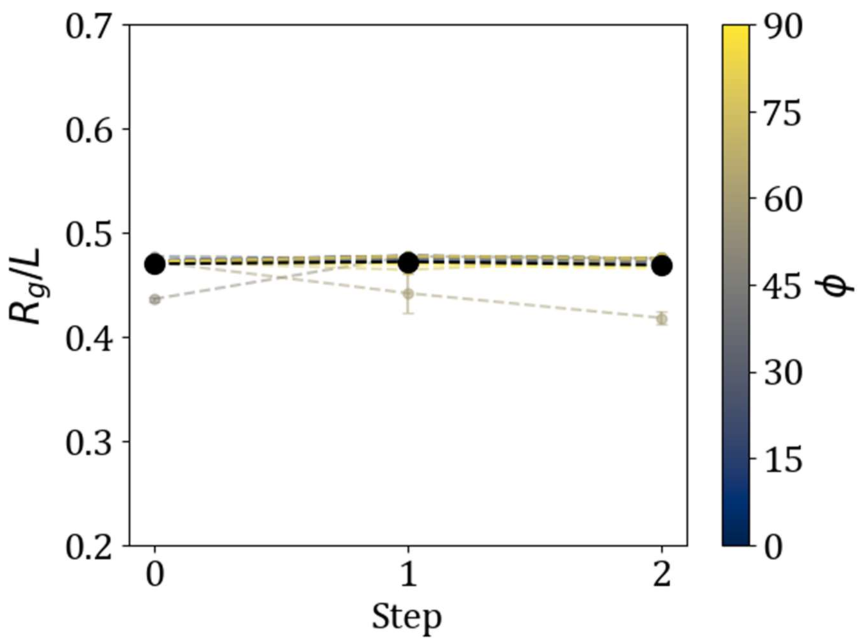

Figure S02.04: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

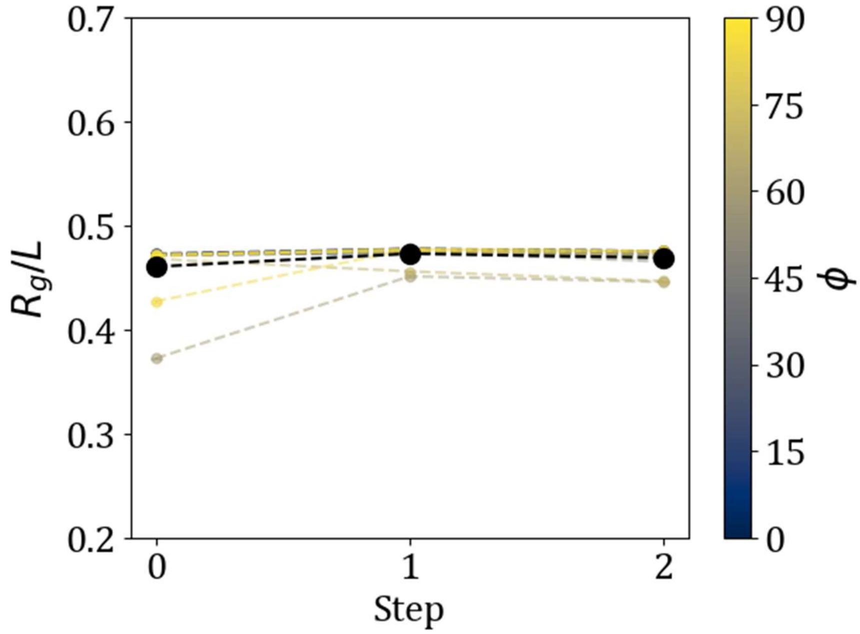

Figure S02.05: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

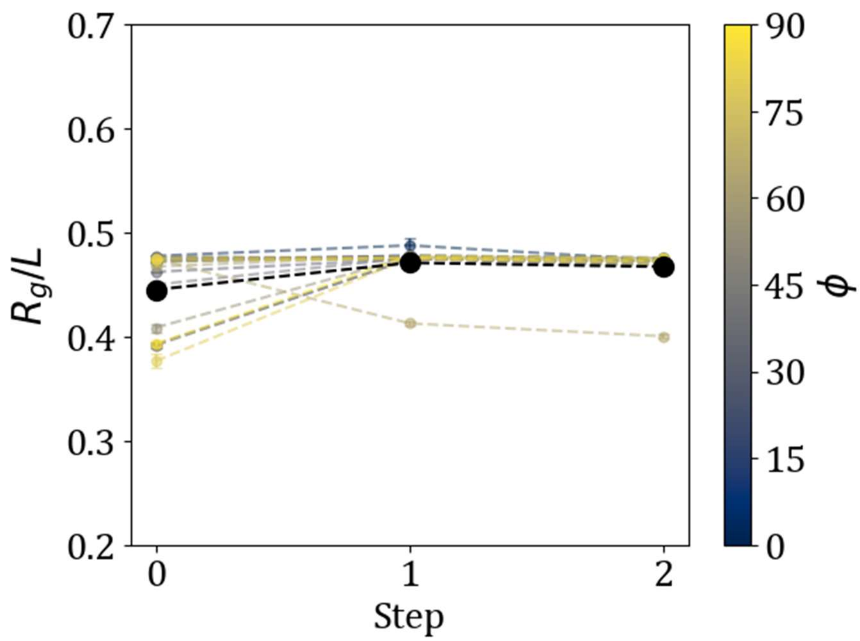

Figure S02.06: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

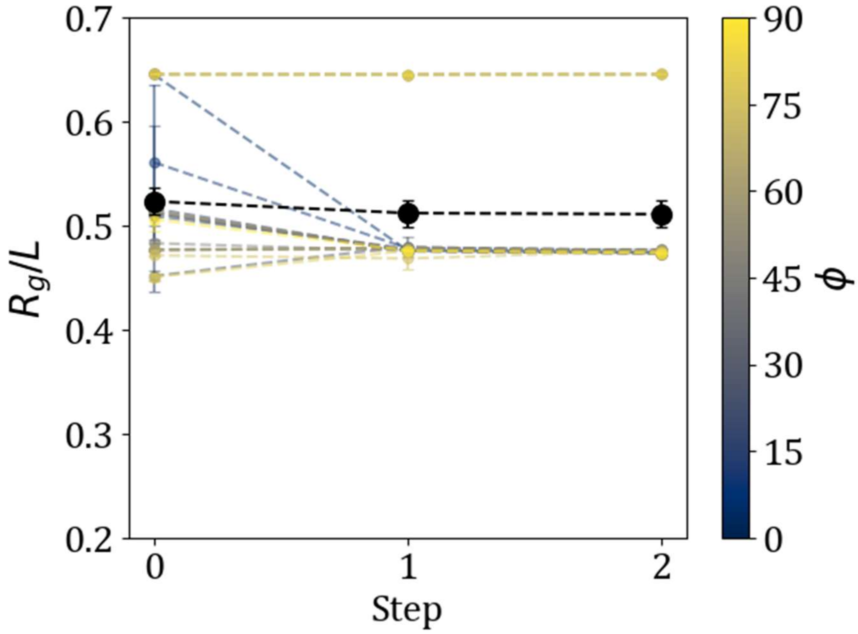

Figure S02.07: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

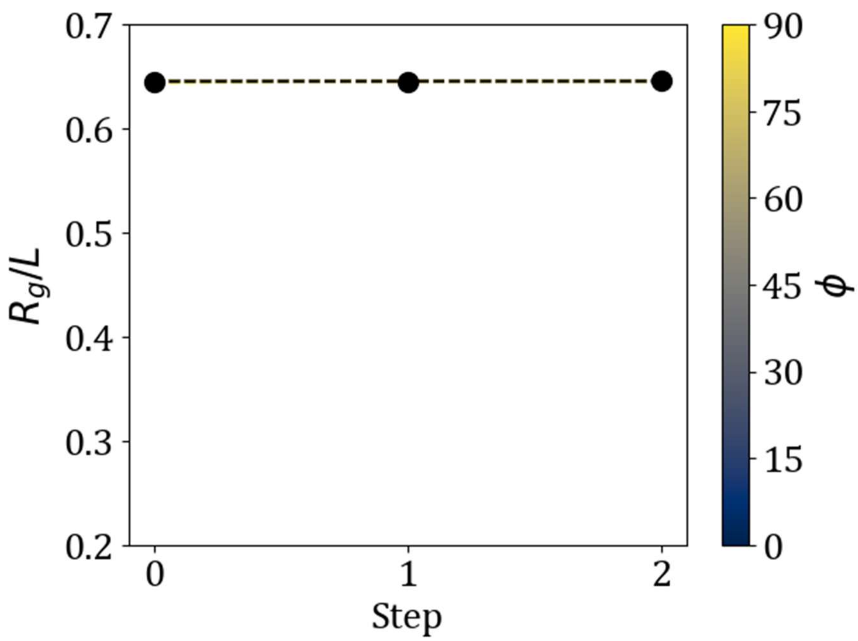

Figure S02.08: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

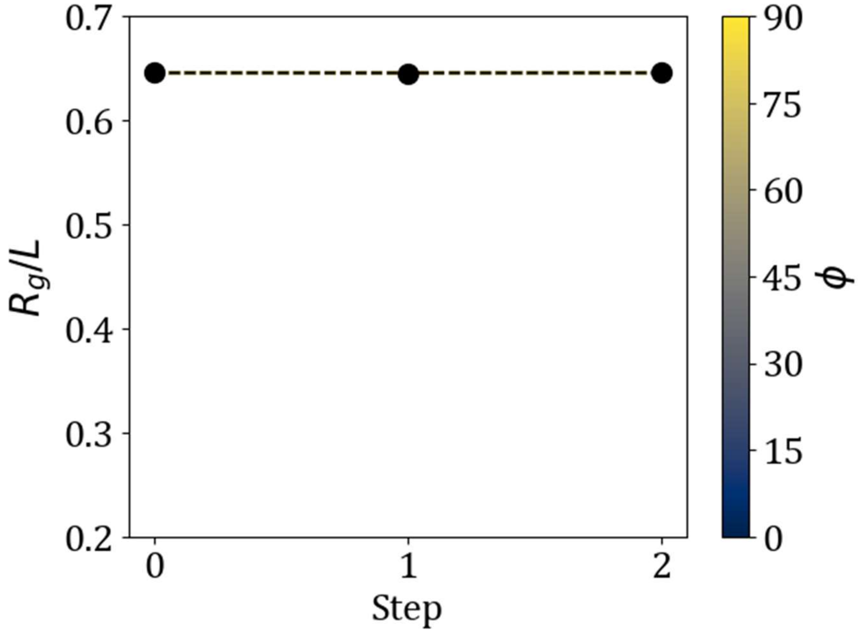

Figure S02.09: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

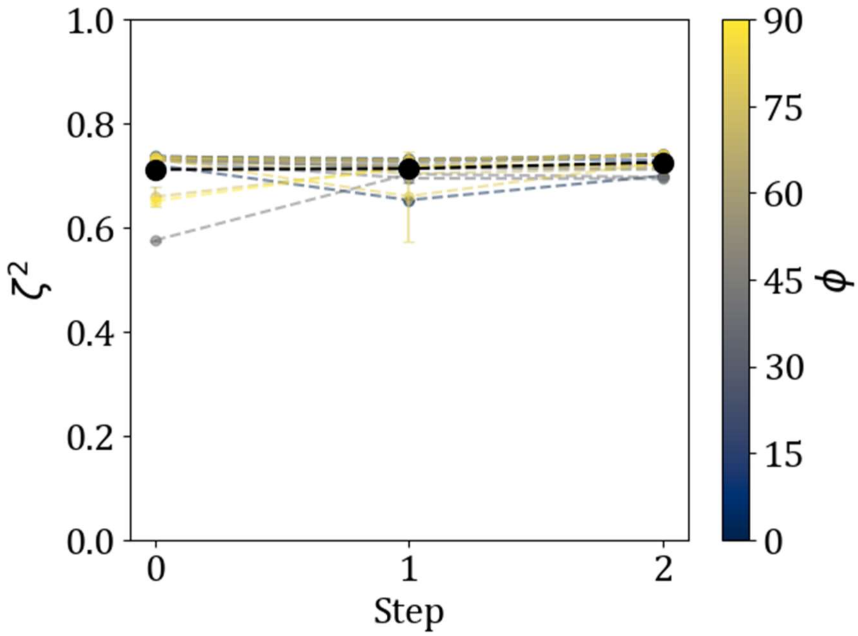

Figure S02.010: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

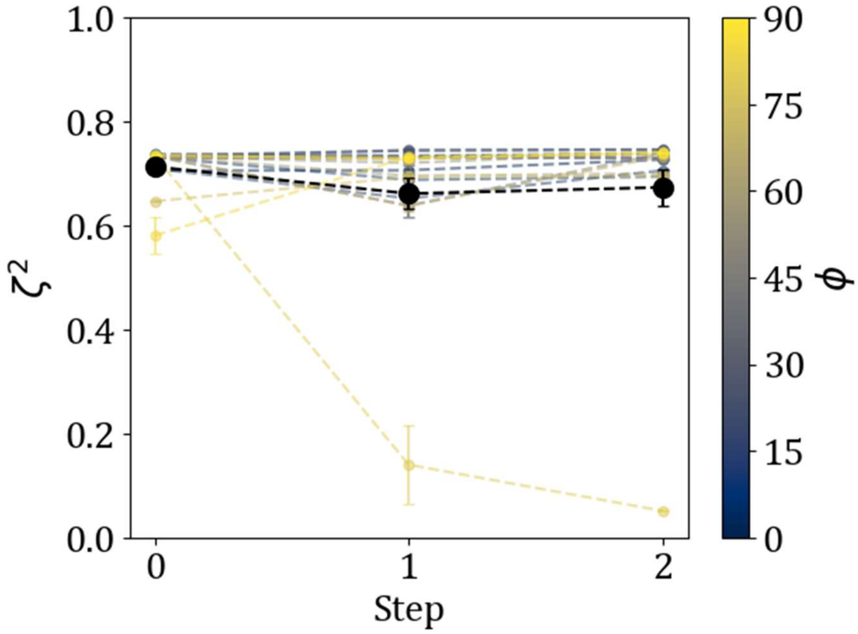

Figure S02.11: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

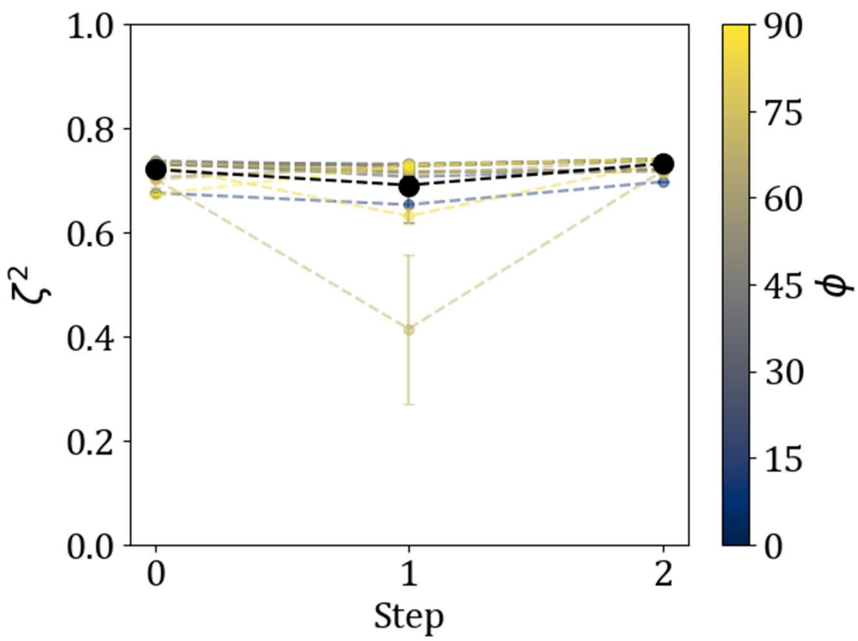

Figure S02.12: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

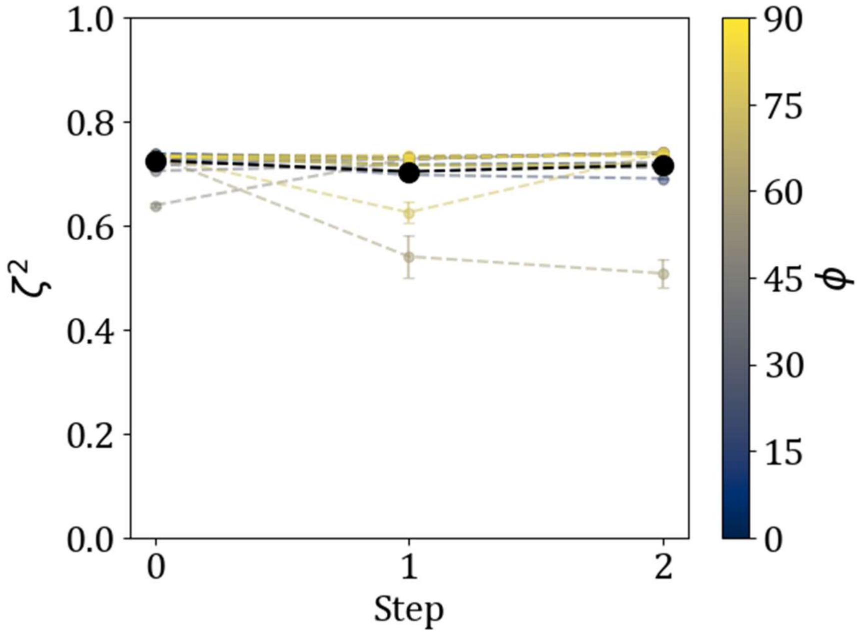

Figure S02.13: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

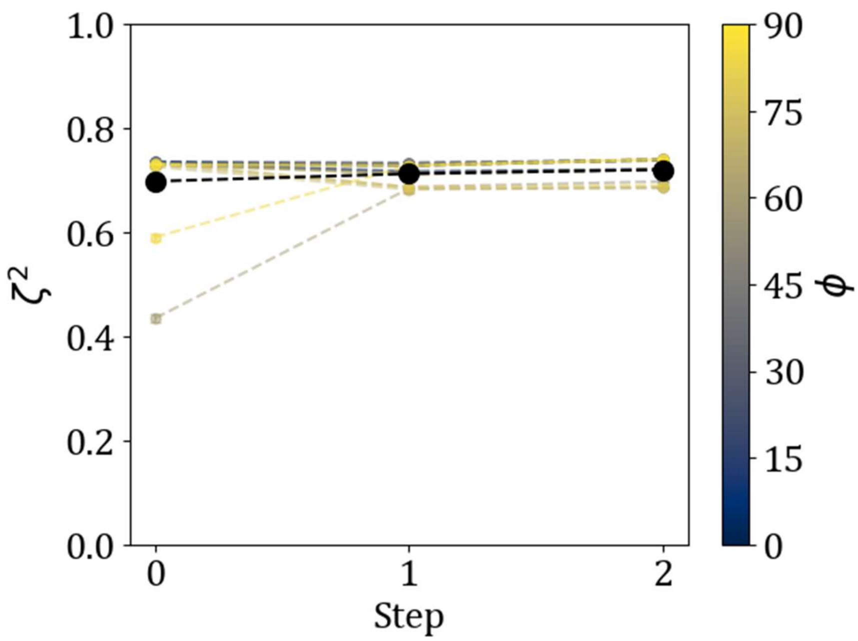

Figure S02.14: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

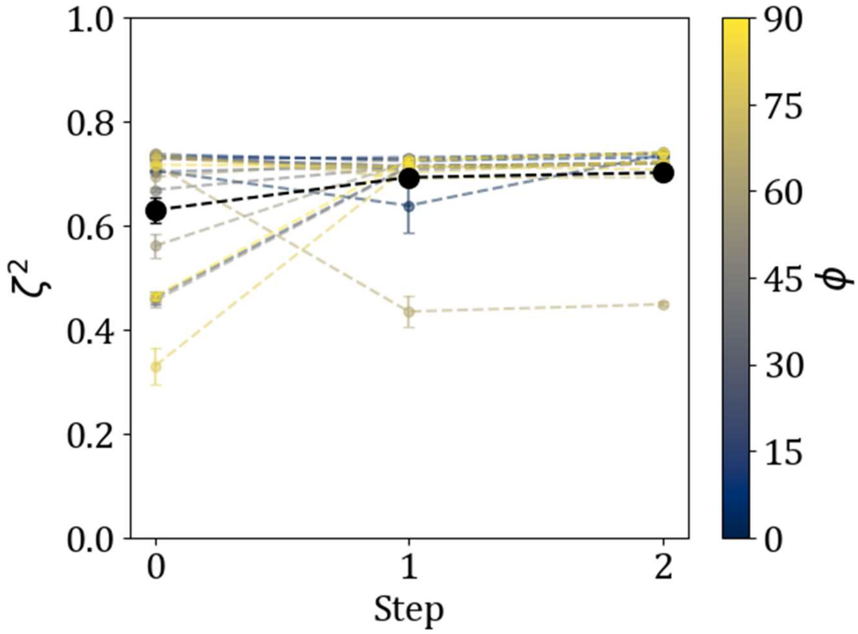

Figure S02.15: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

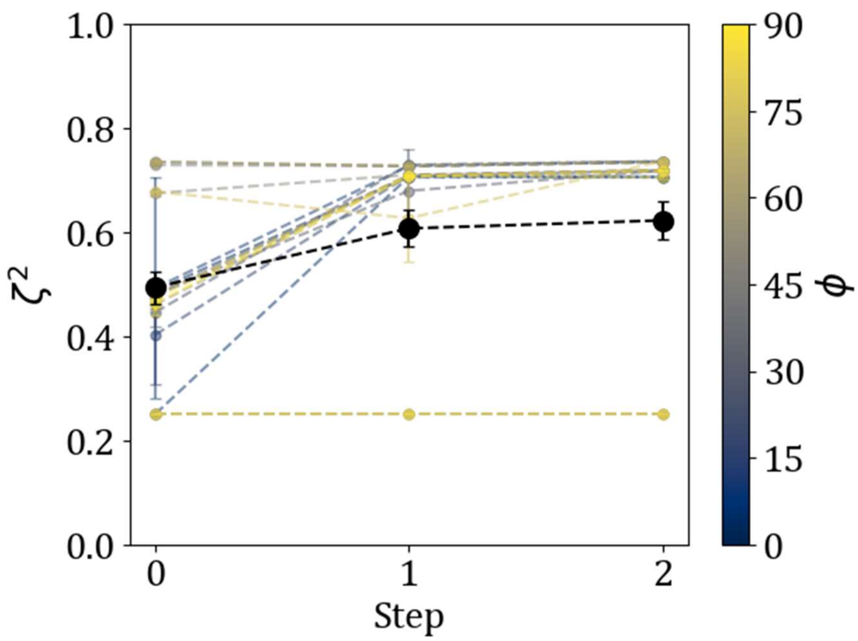

Figure S02.16: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

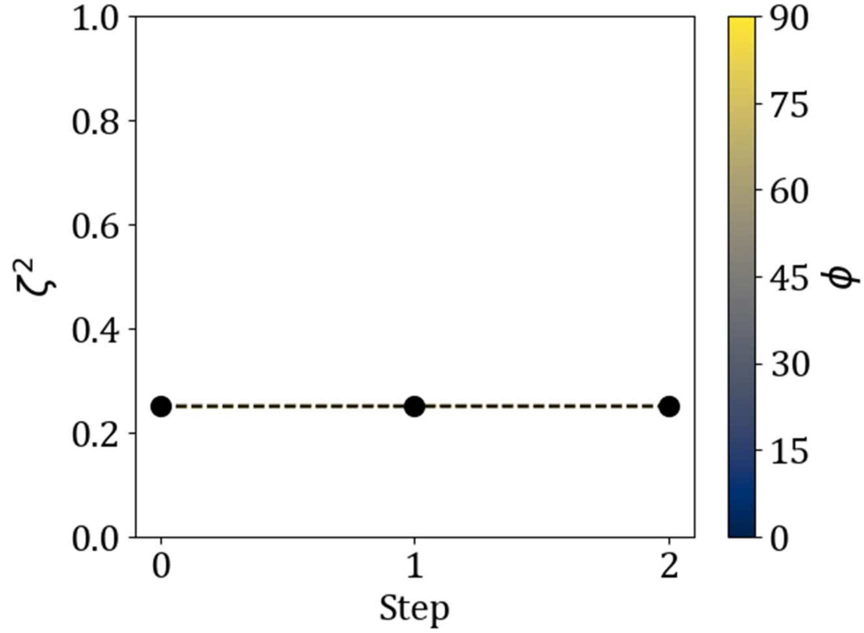

Figure S02.17: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

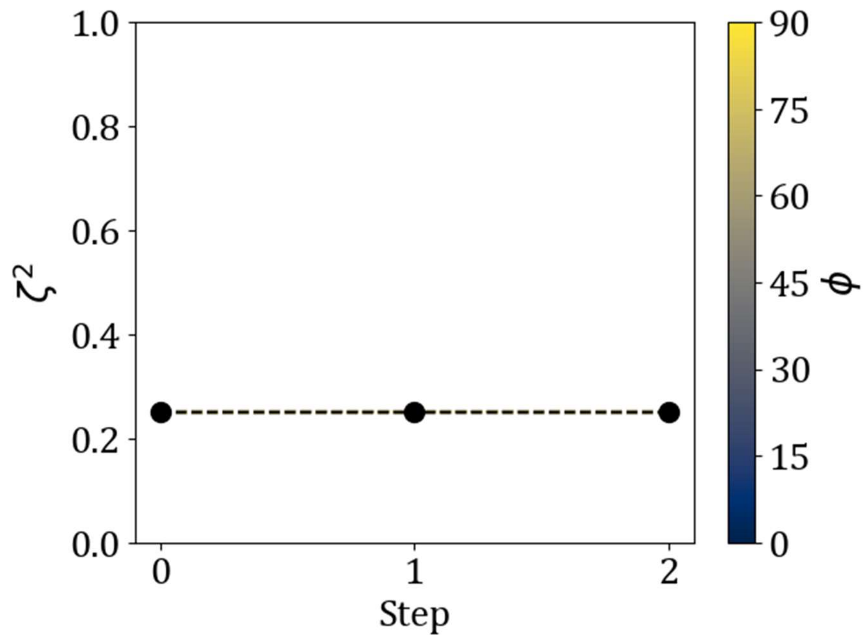

Figure S02.18: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

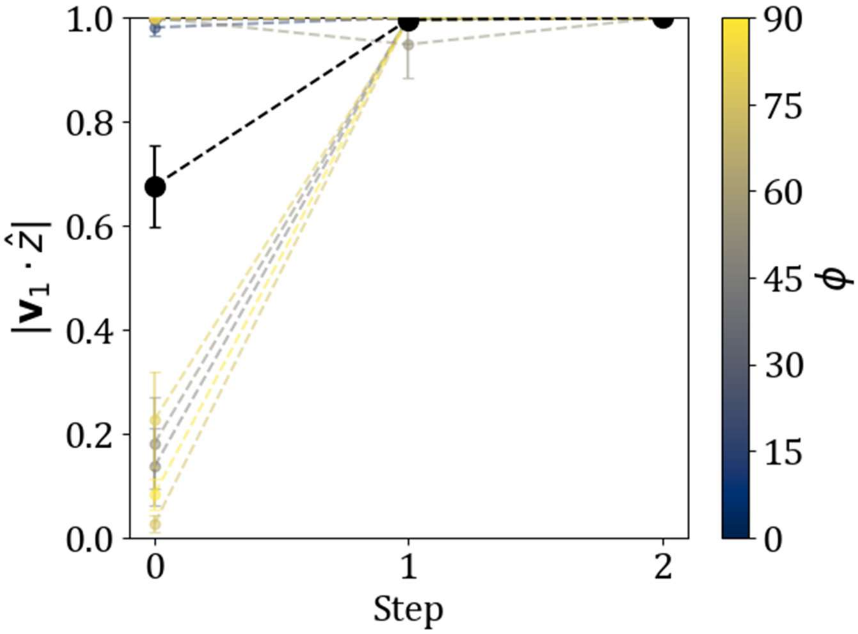

Figure S02.19: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

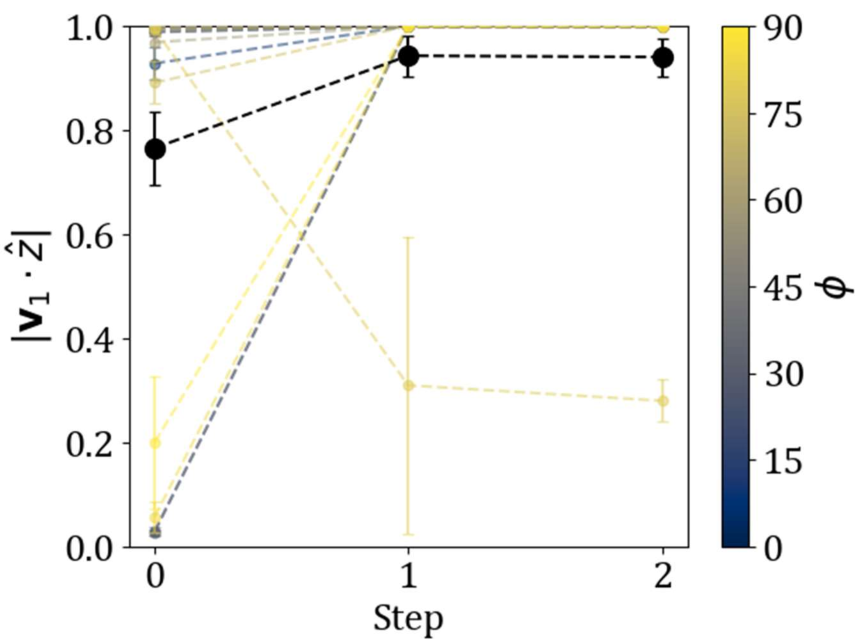

Figure S02.20: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

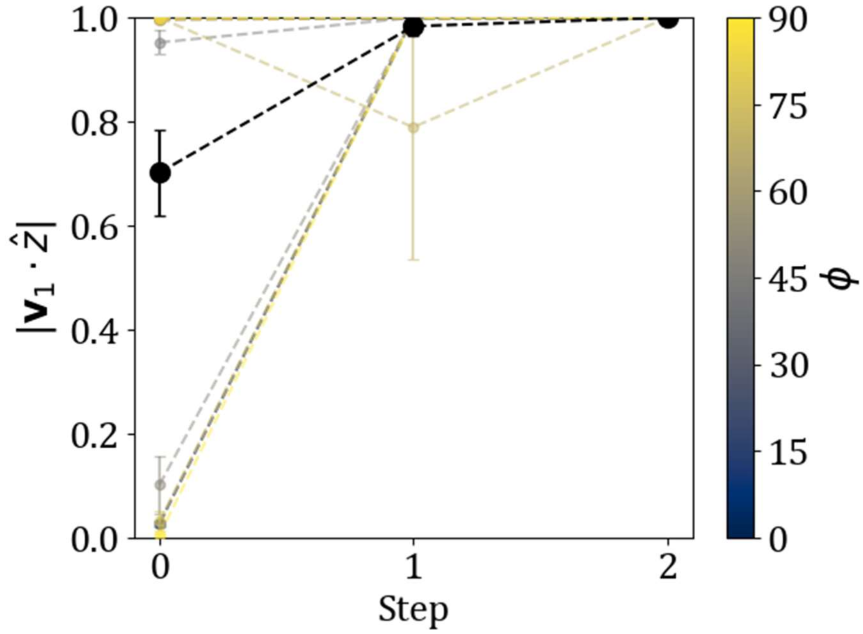

Figure S02.21: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

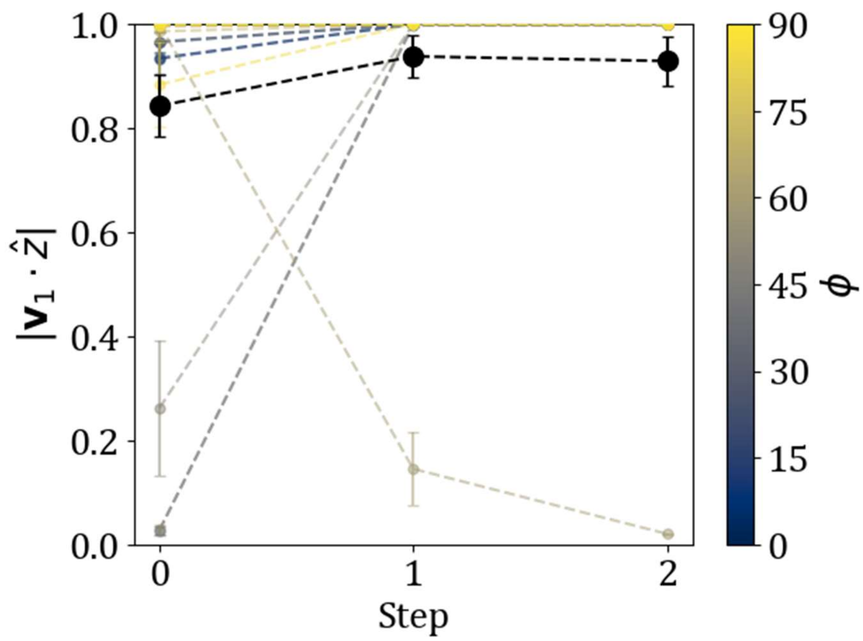

Figure S02.22: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

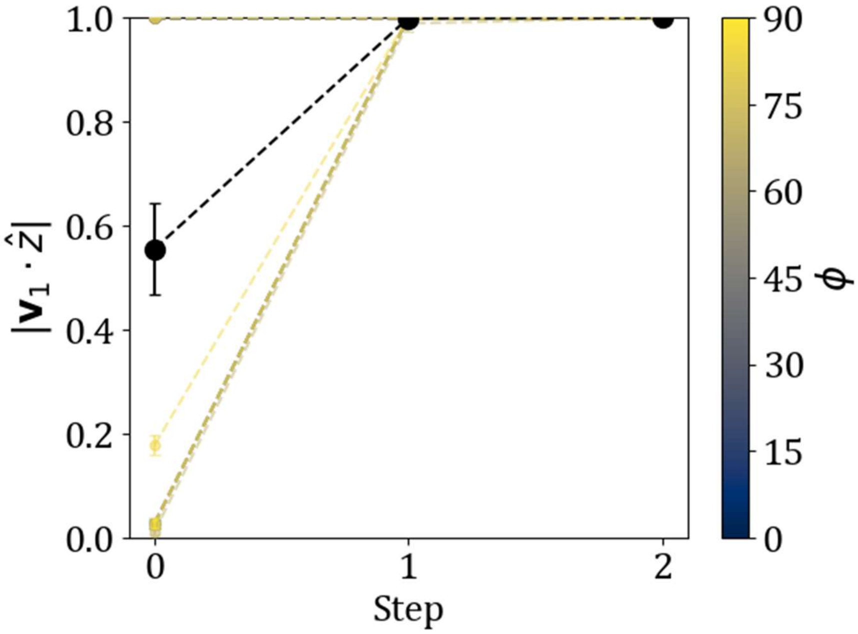

Figure S02.23: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

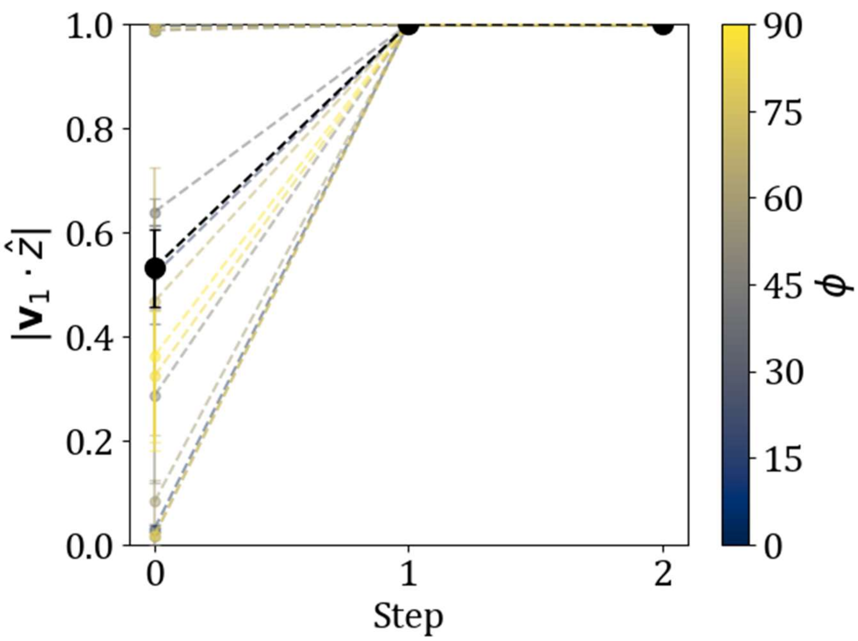

Figure S02.24: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

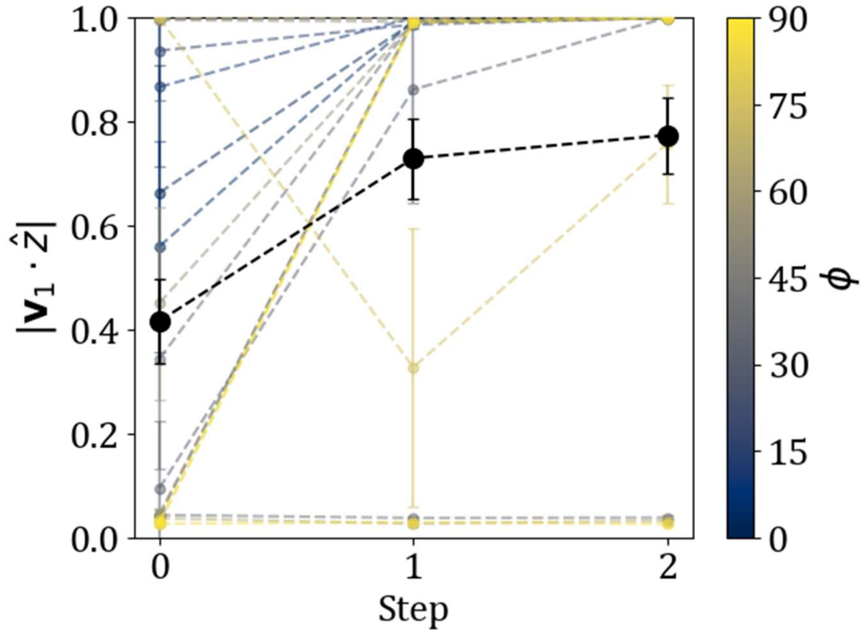

Figure S02.25: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

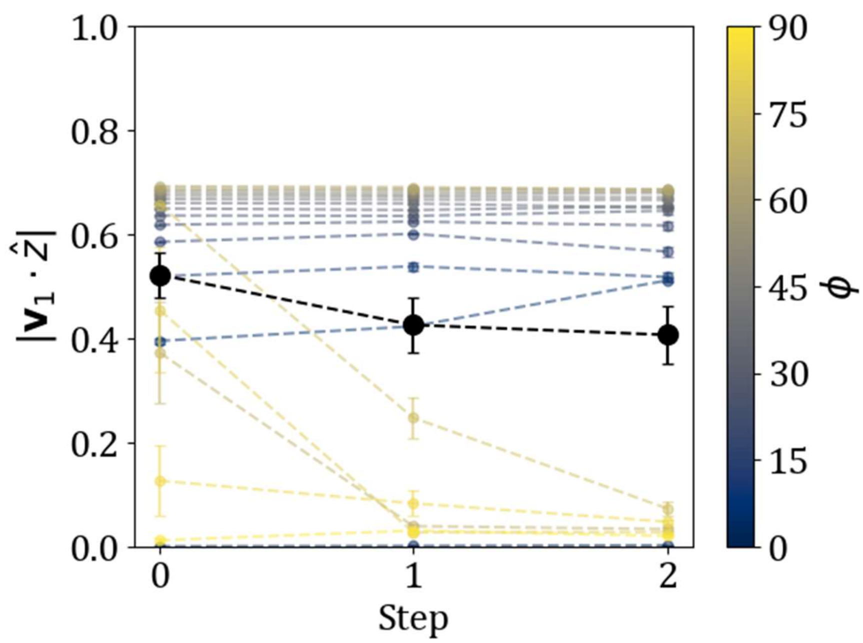

Figure S02.26: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

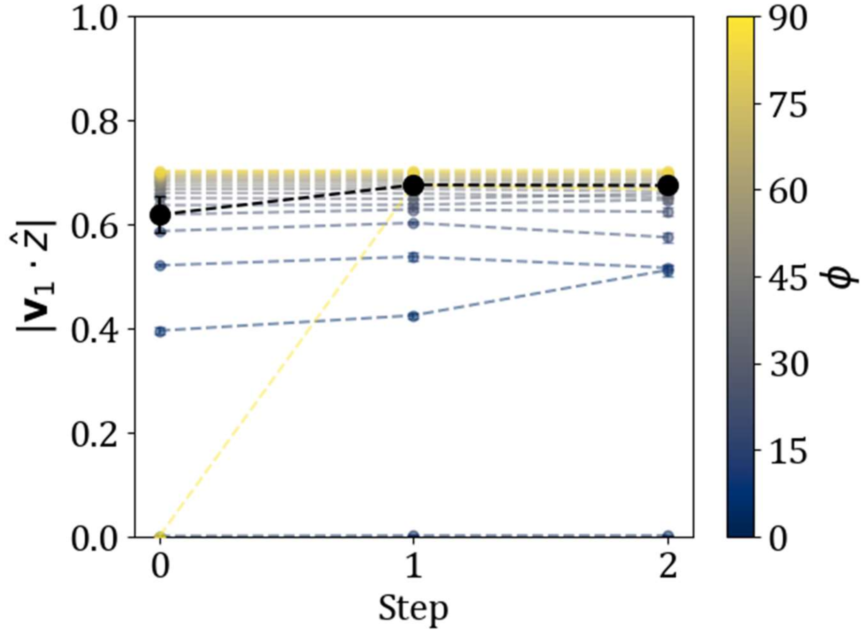

Figure S02.27: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 1 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

SI.03: Summary statistics for all Protocol 2 simulations

In the main article, we show only averages over all initial conditions. Here, we give results for all initial conditions, grouped by protocol and the value of K.

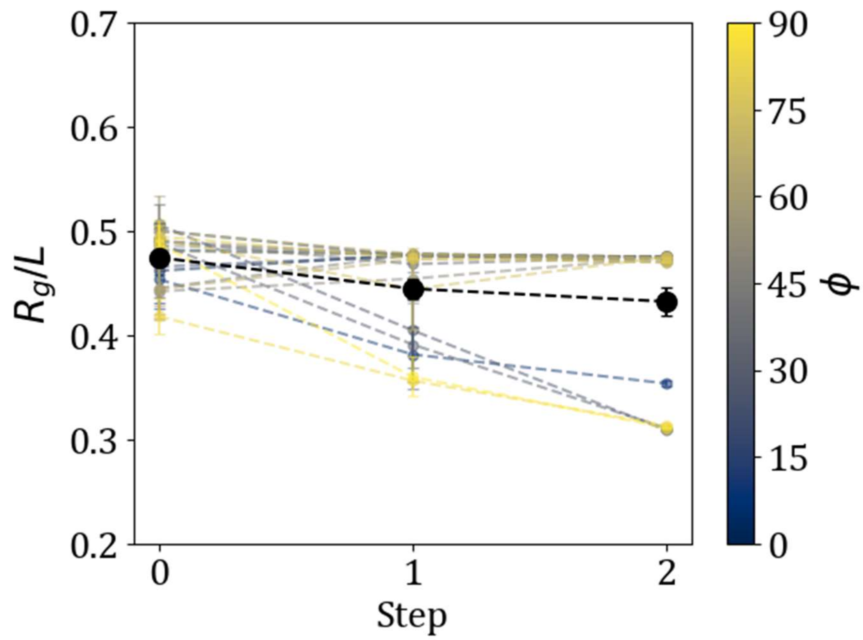

Figure S03.01: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

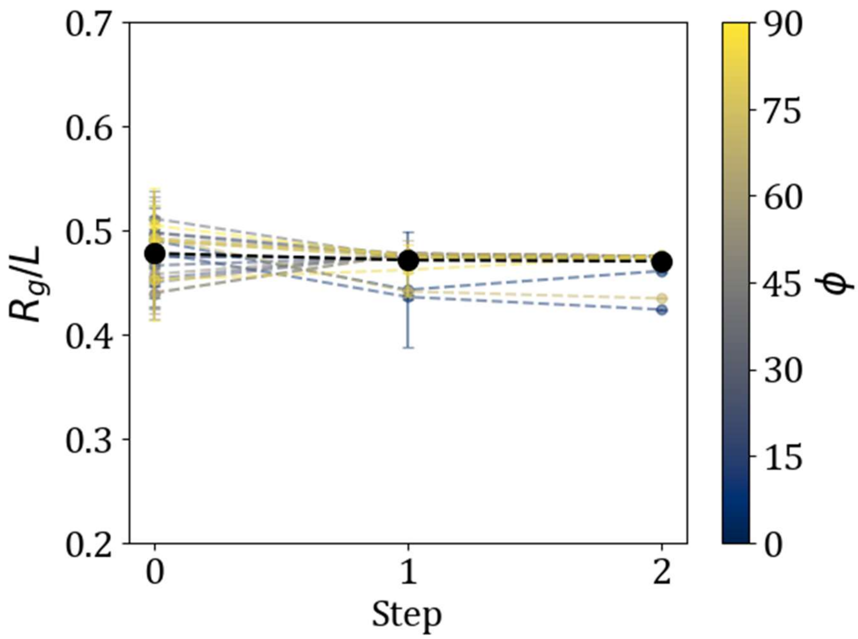

Figure S03.02: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

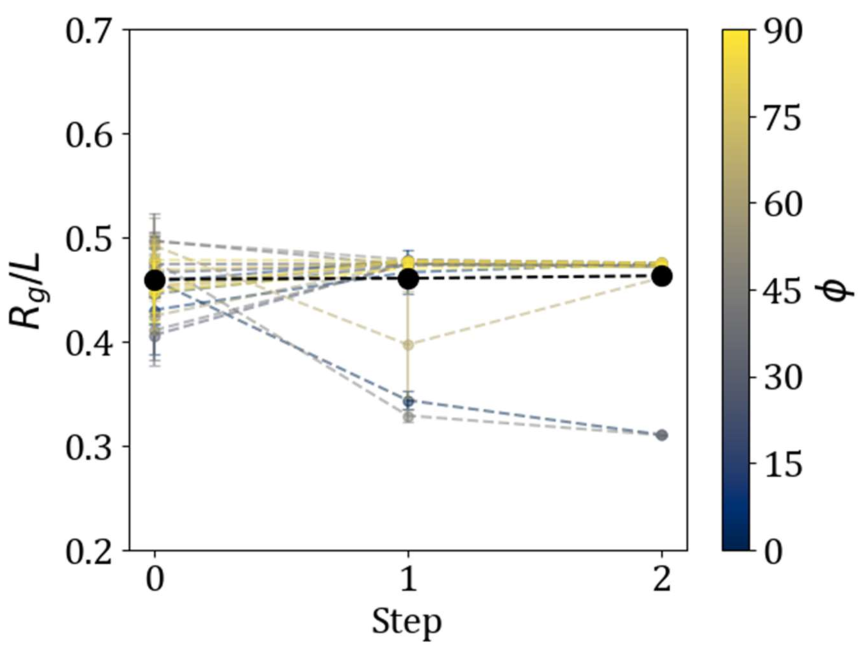

Figure S03.03: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

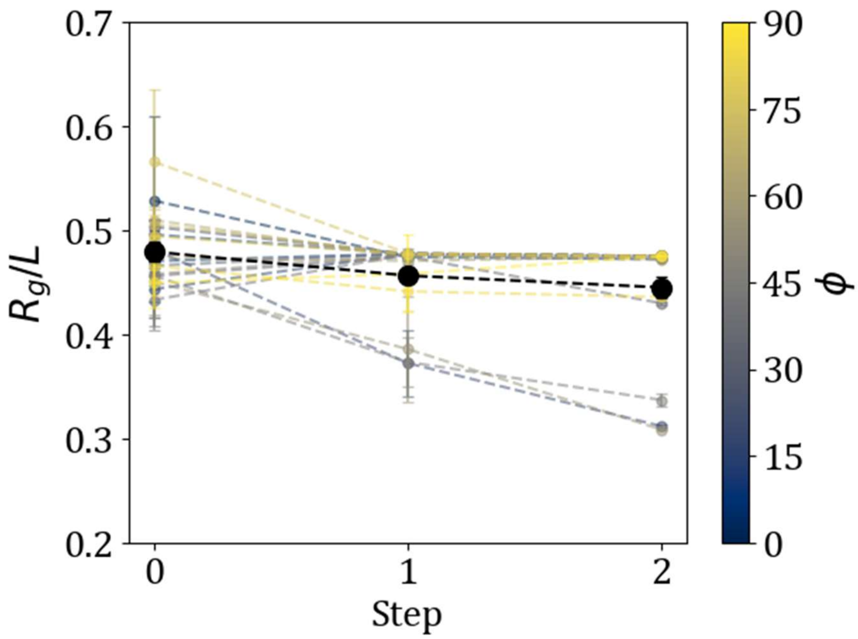

Figure S03.04: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

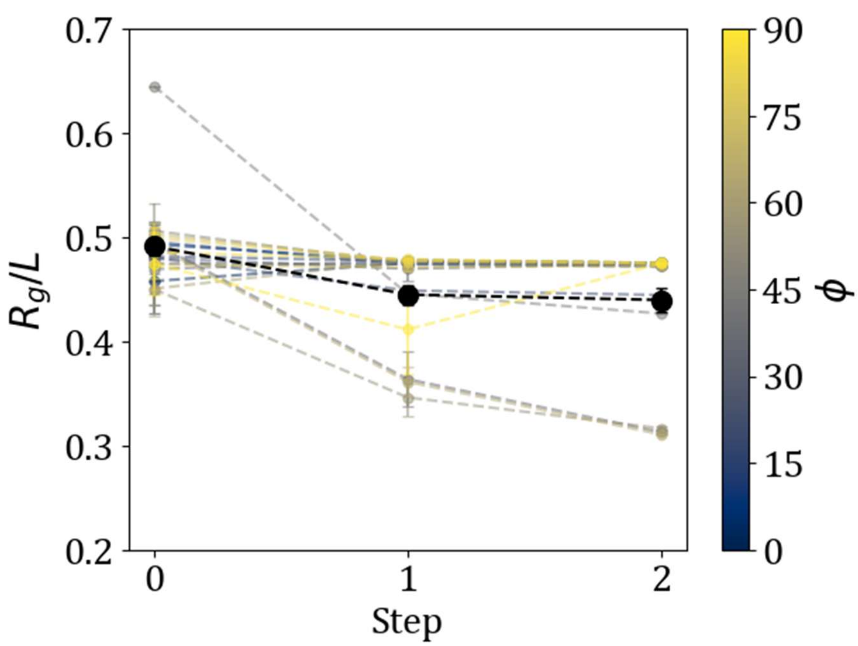

Figure S03.05: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

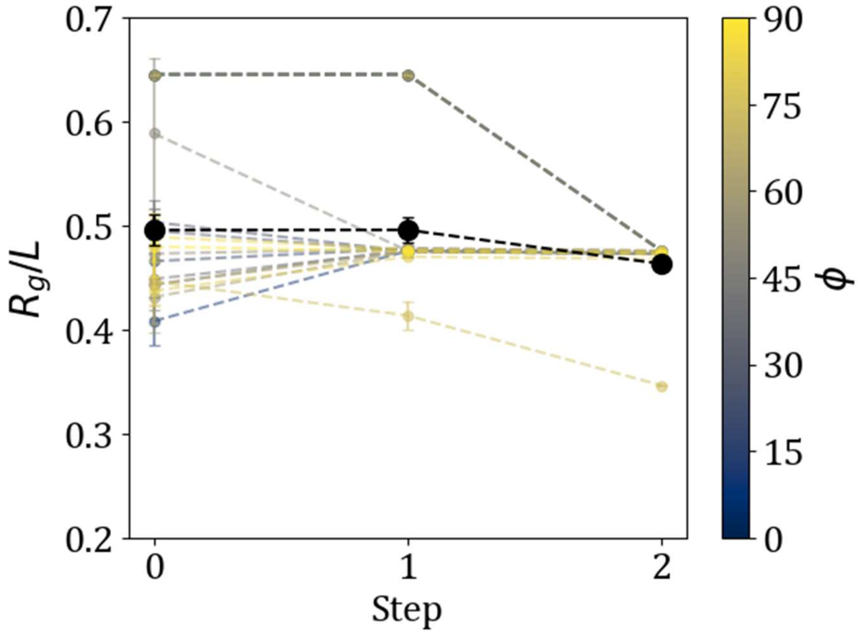

Figure S03.06: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

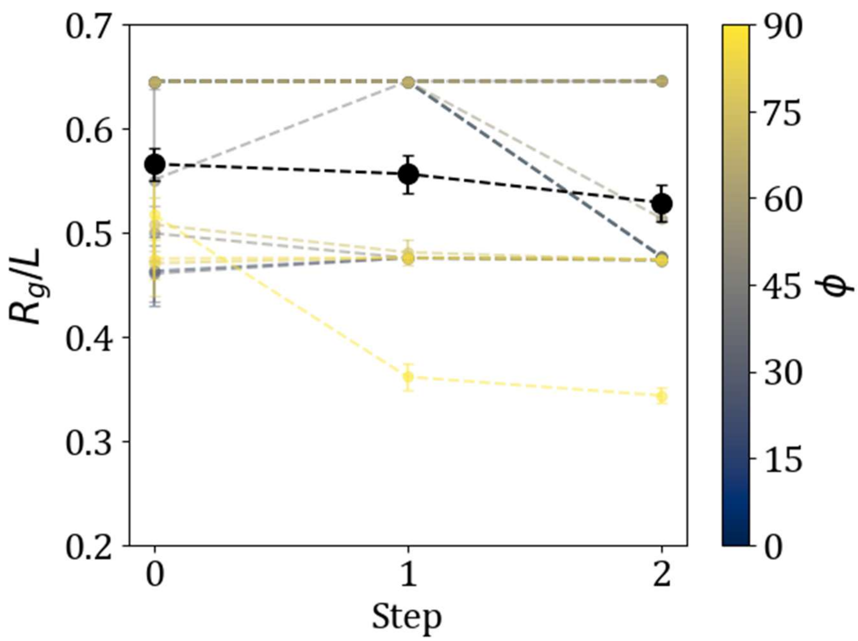

Figure S03.07: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

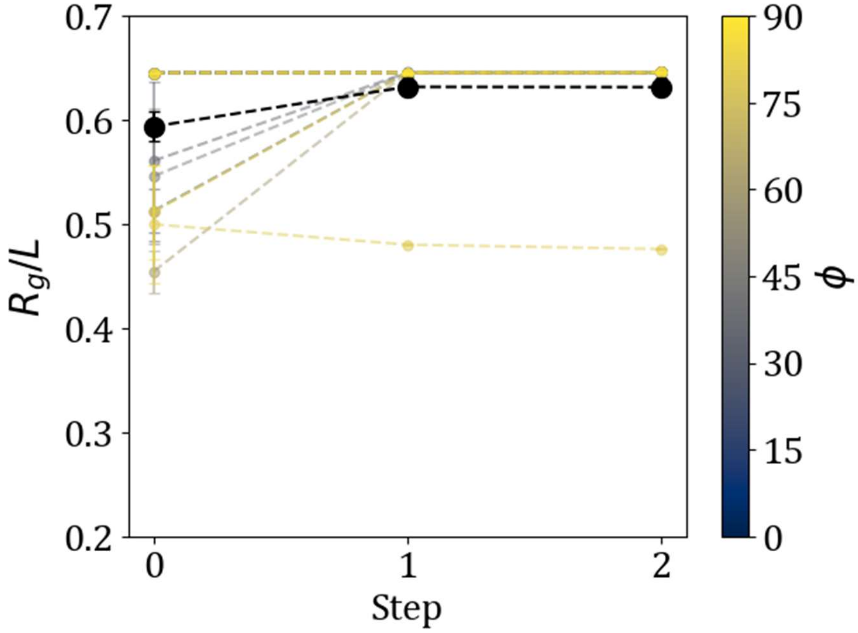

Figure S03.08: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

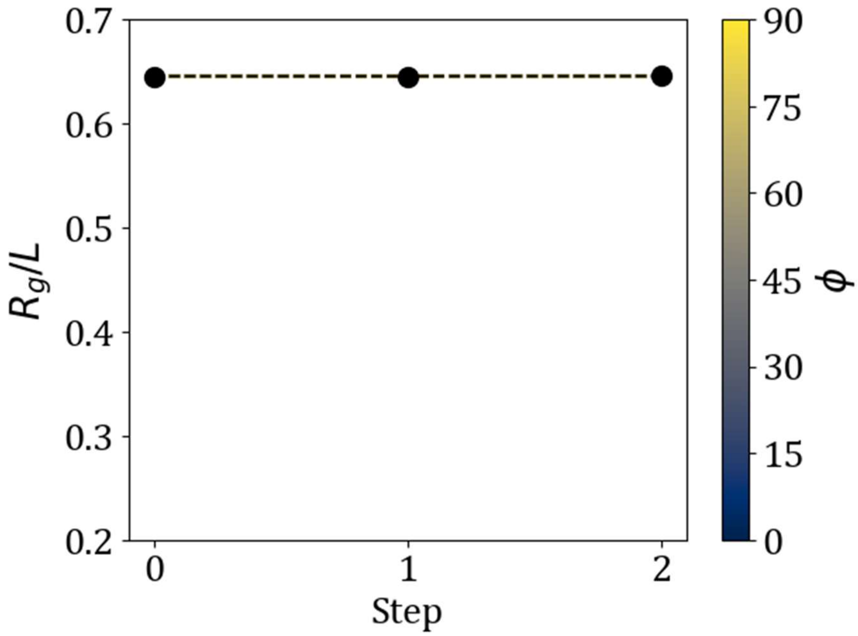

Figure S03.09: Radius of gyration averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

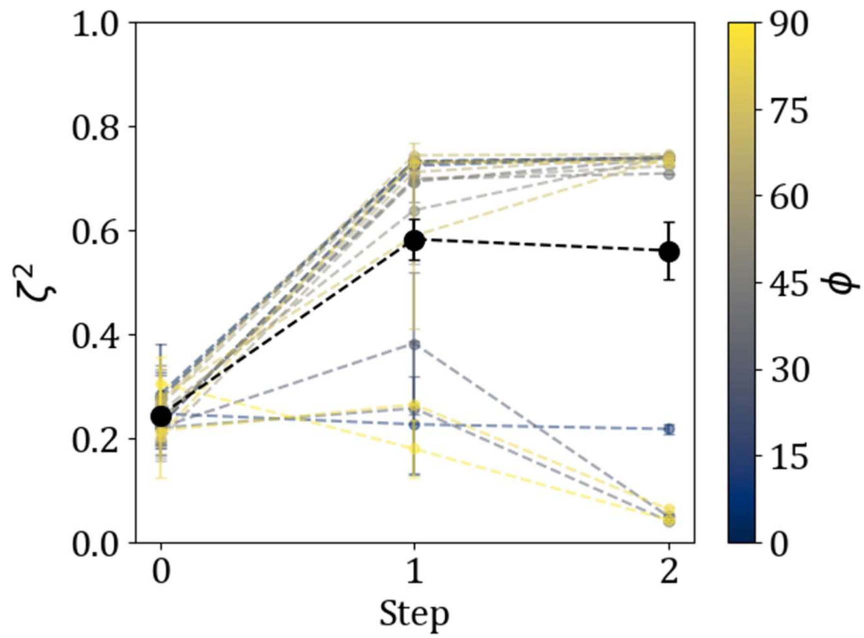

Figure S03.010: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

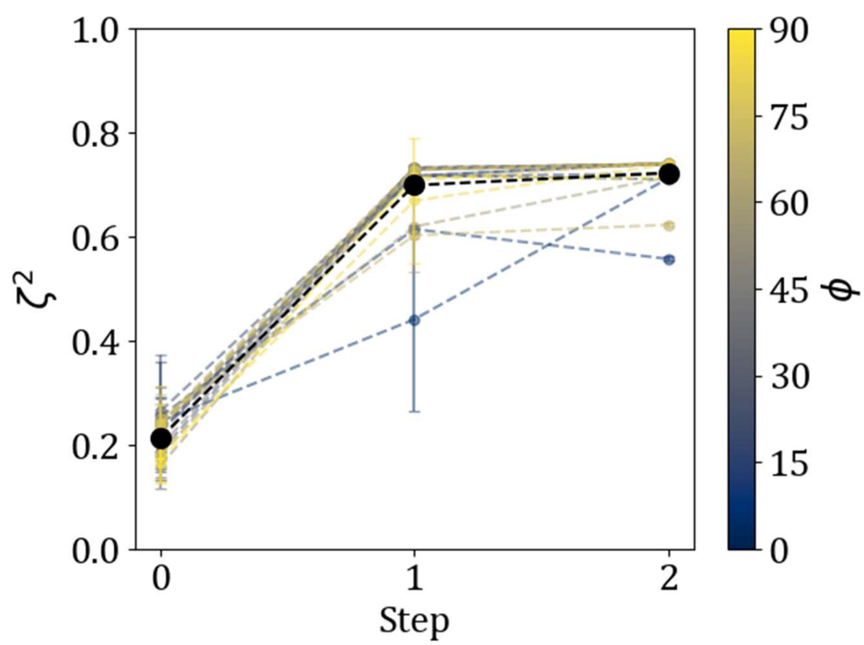

Figure S03.11: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

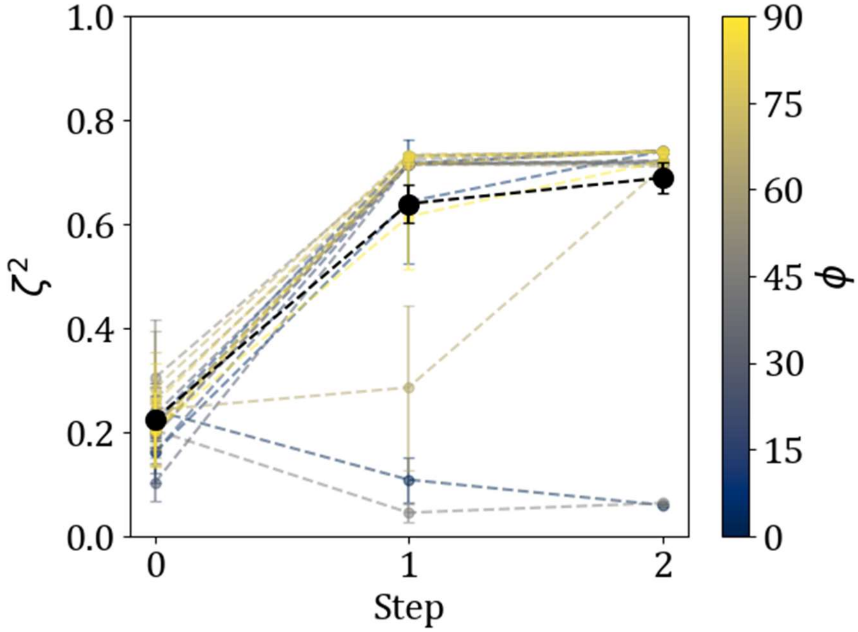

Figure S03.12: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

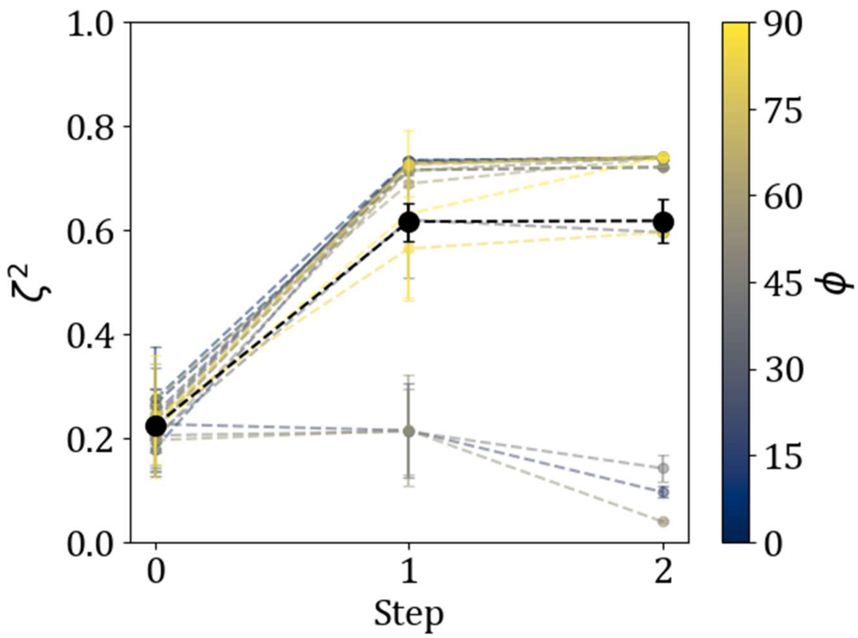

Figure S03.13: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

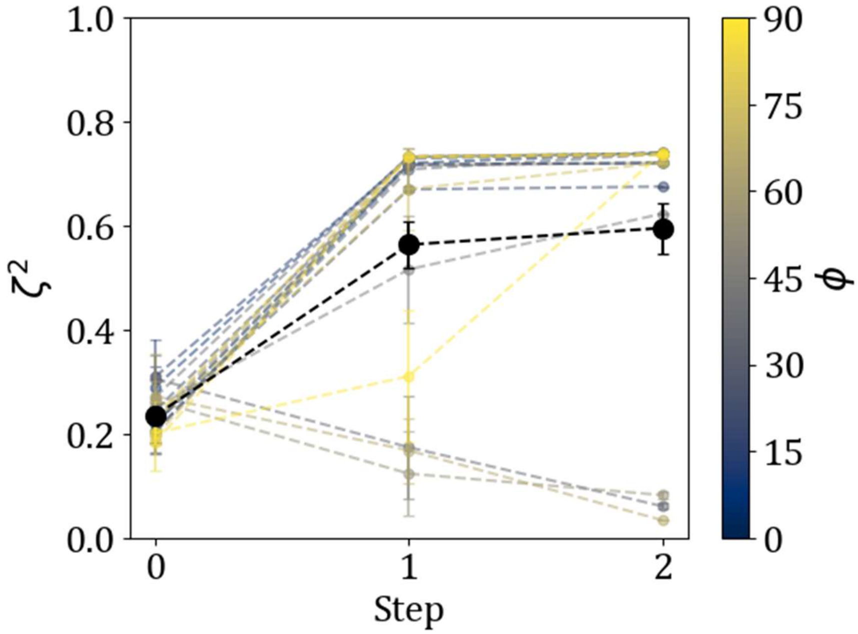

Figure S03.14: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

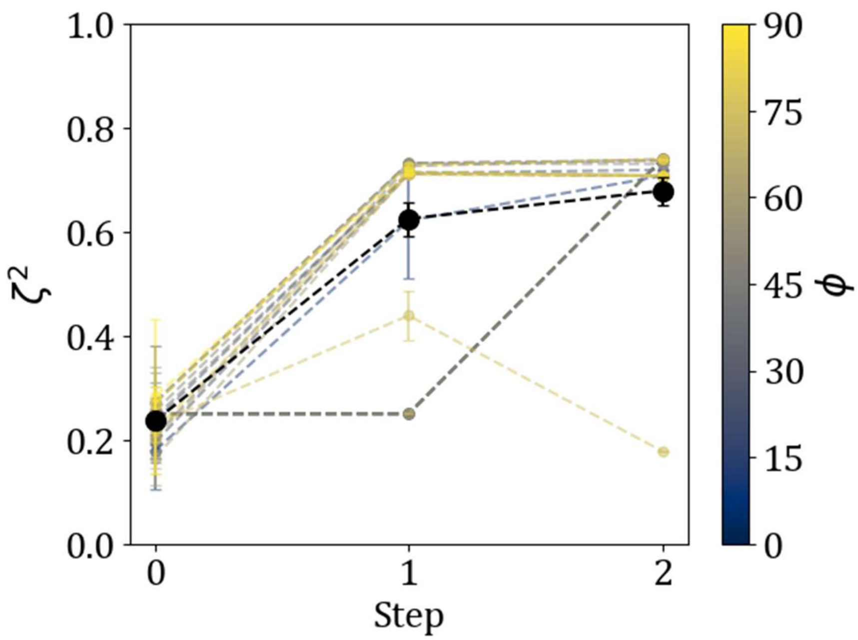

Figure S03.15: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

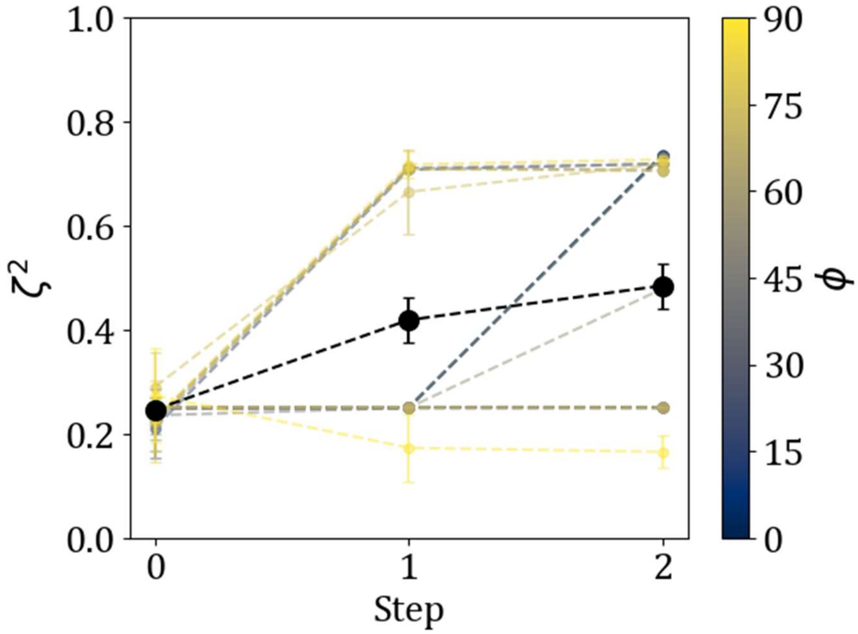

Figure S03.16: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

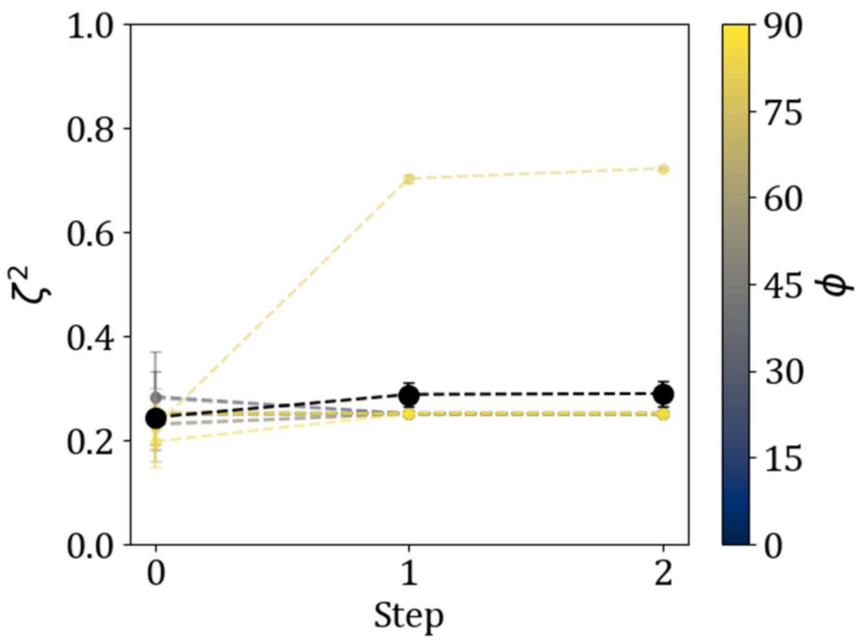

Figure S03.17: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

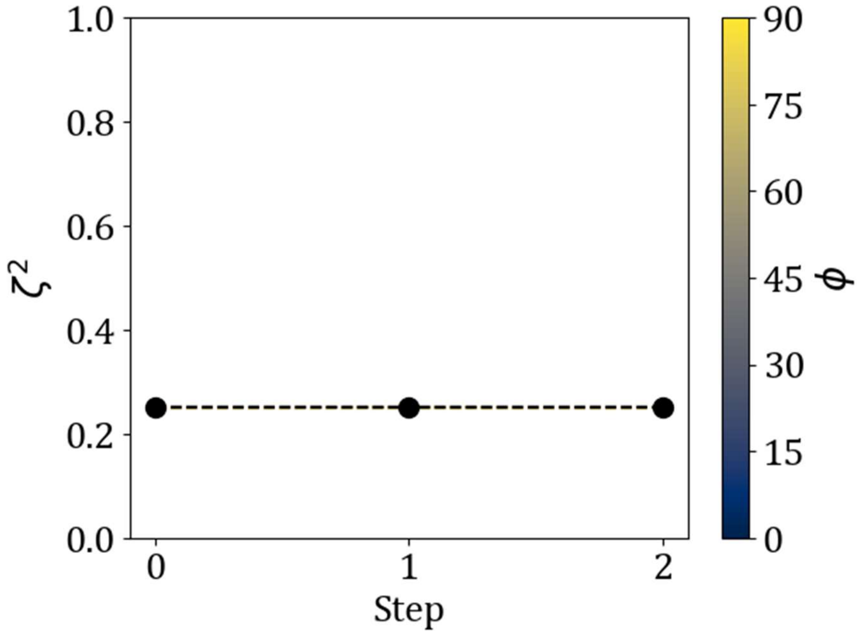

Figure S03.18: Relative shape anisotropy averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

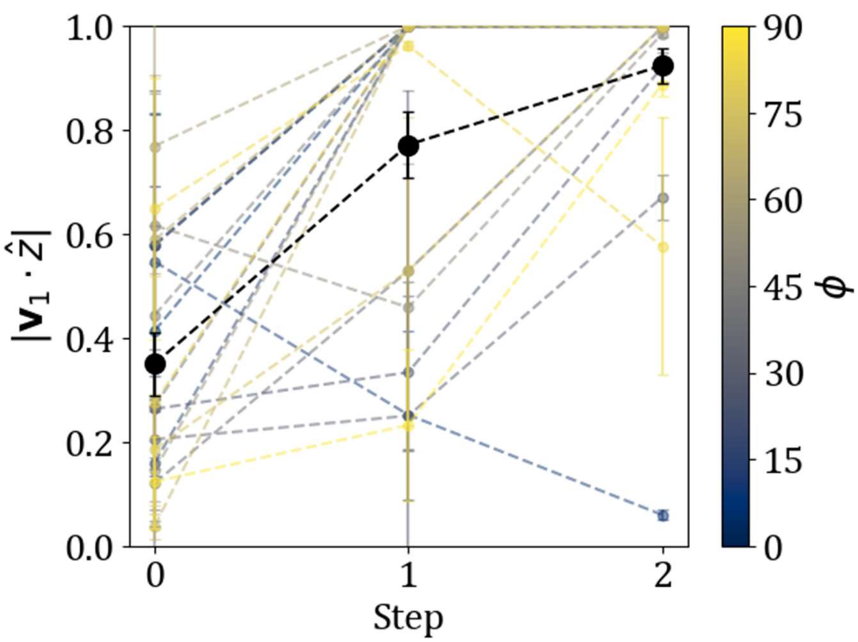

Figure S03.19: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.003$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

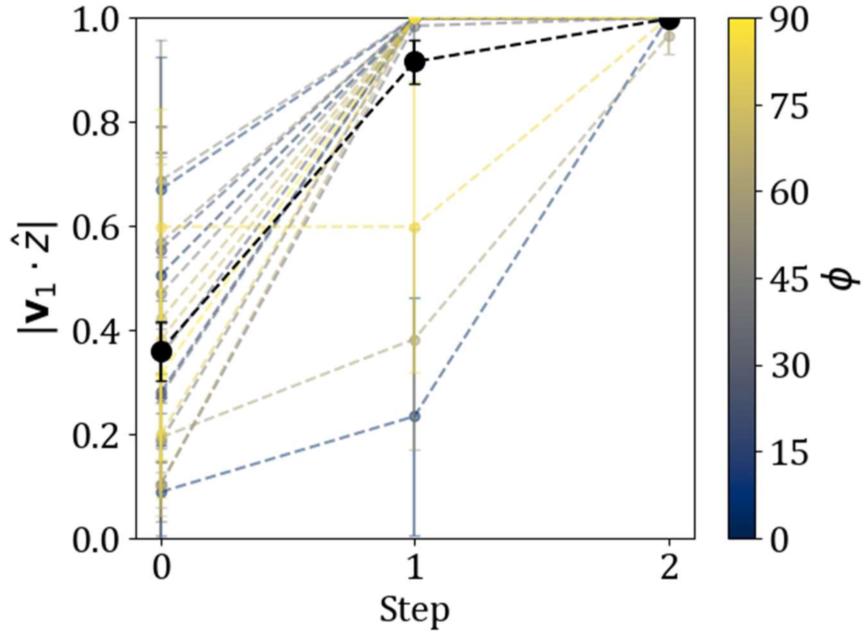

Figure S03.20: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.01$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

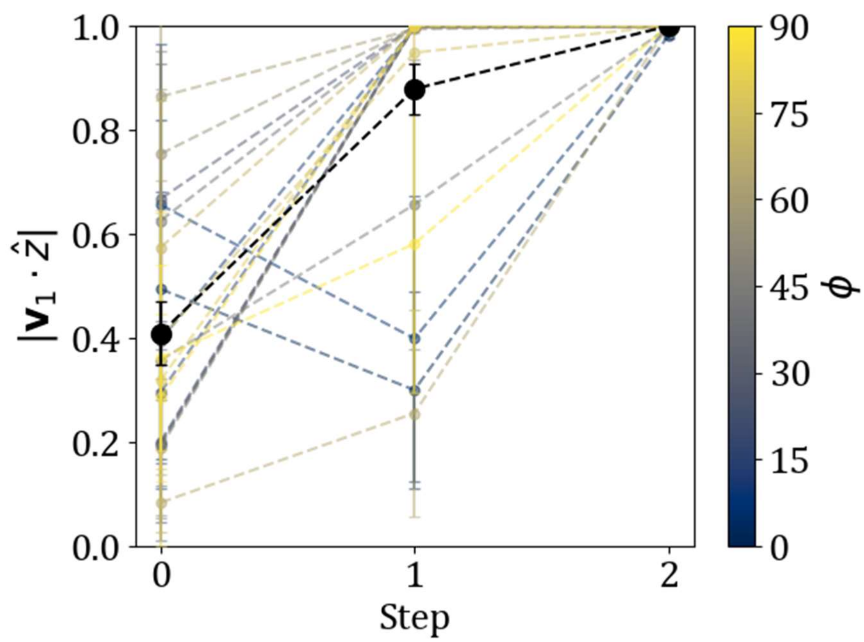

Figure S03.21: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.03$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

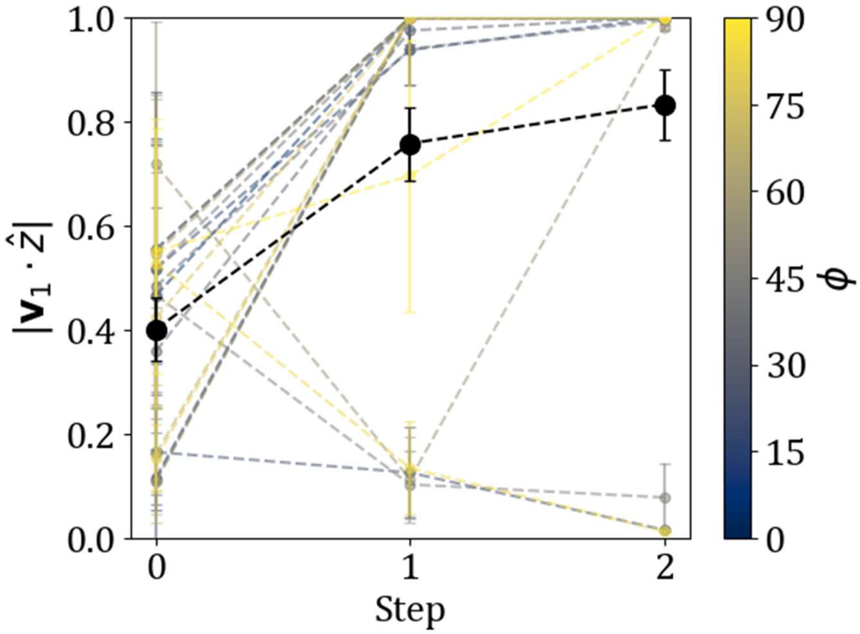

Figure S03.22: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.1$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

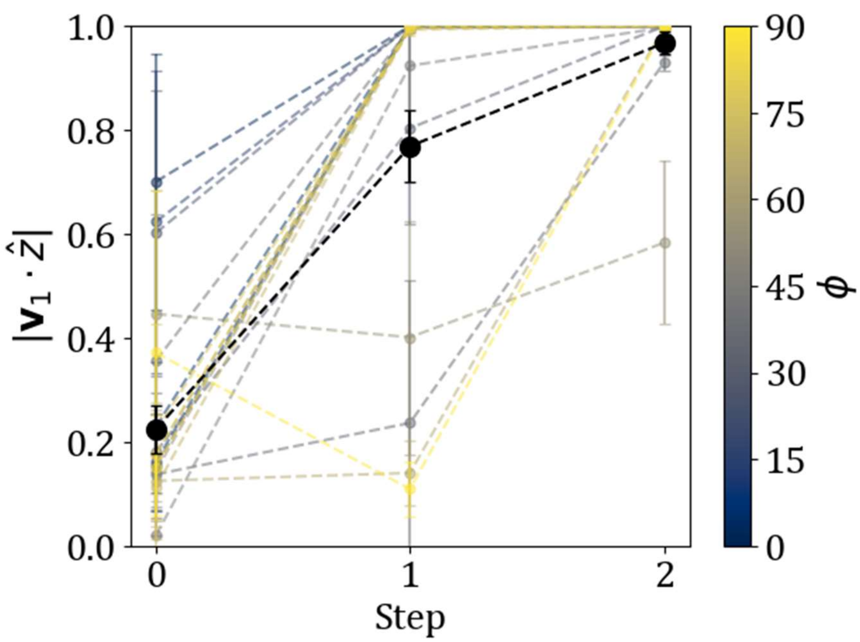

Figure S03.23: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 0.3$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

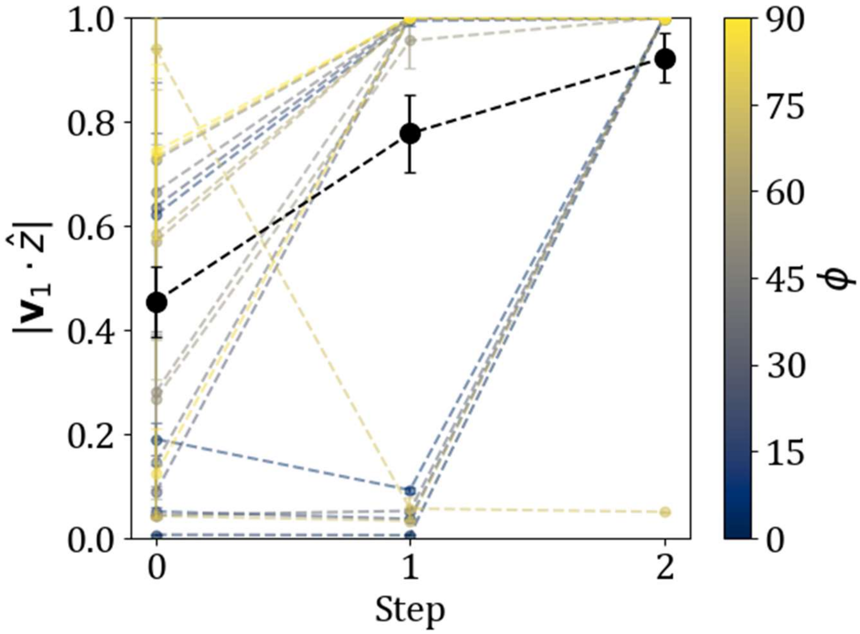

Figure S03.24: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 1.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

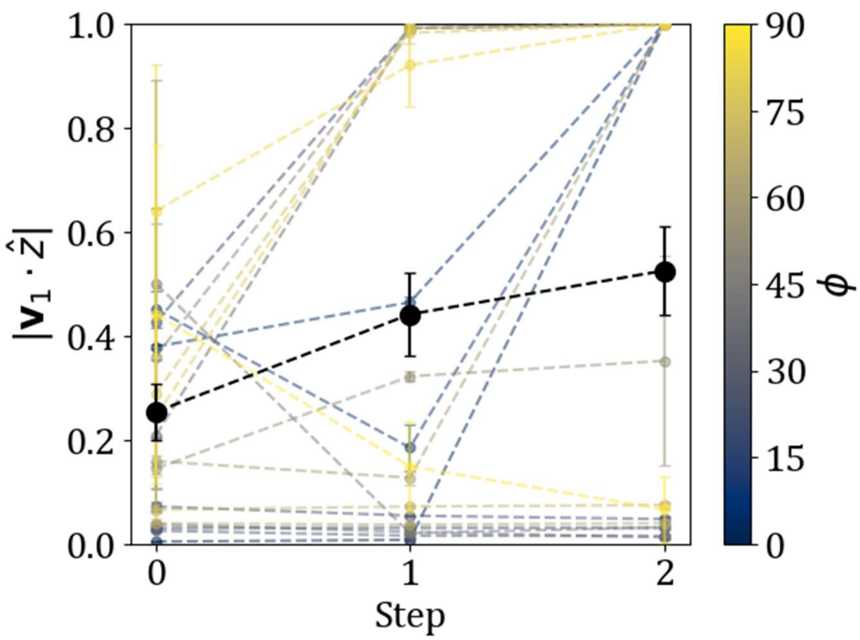

Figure S03.25: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 3.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

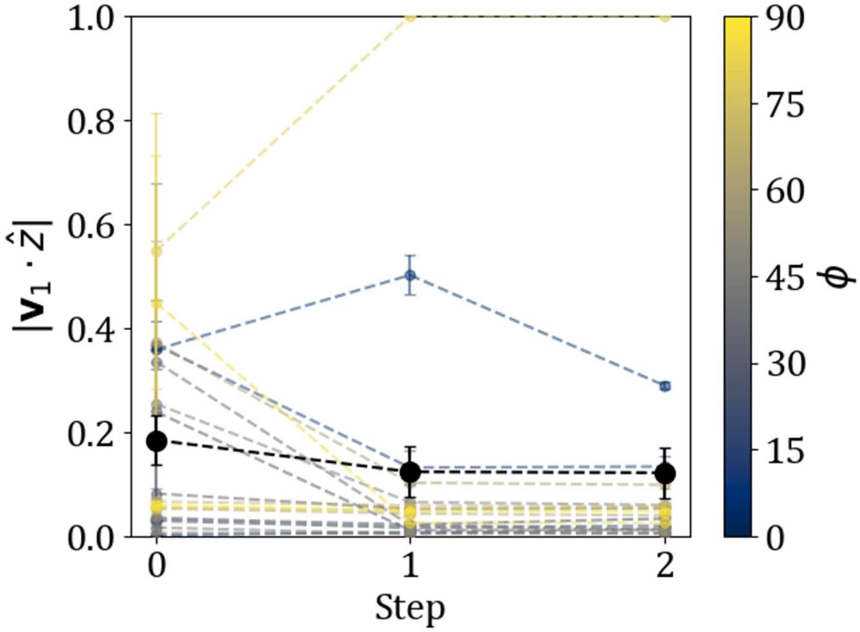

Figure S03.26: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 10.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

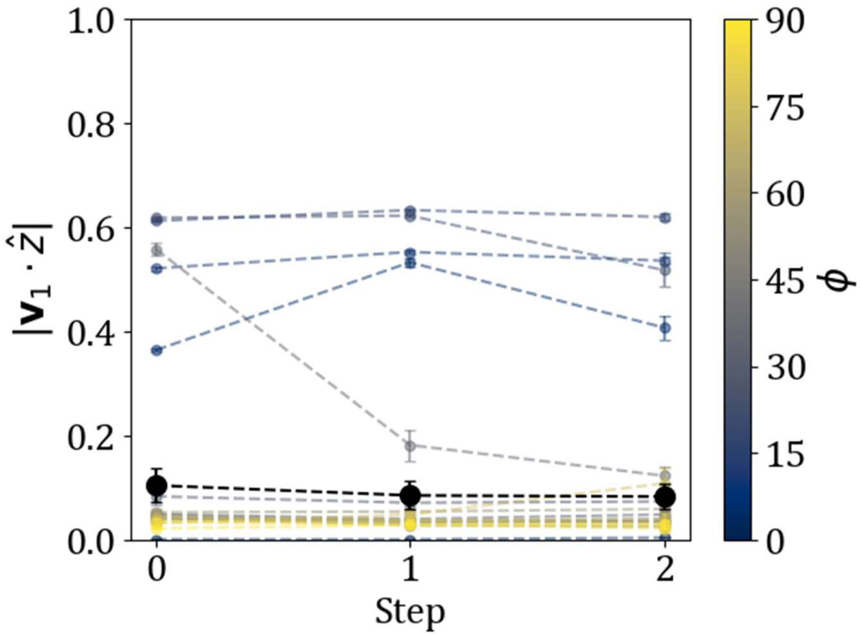

Figure S03.27: Alignment averaged over every $100\dot{\gamma}t$ for the last $500\dot{\gamma}t$ of each step for Protocol 2 with $K = 30.0$. Black dots show the weighted average over all initial conditions. Dotted lines are drawn to guide the eye.

SI.04: References

[1]Funkenbusch, W. T., Silmore, K. S., & Doyle, P. S. (2024). Dynamics of a self-interacting sheet in shear flow. *Soft Matter*.

[2]Stukowski, A. (2009). Visualization and analysis of atomistic simulation data with OVITO–the Open Visualization Tool. *Modelling and simulation in materials science and engineering*, *18*(1), 015012.