# **Electronic Supporting Information**

# Dipolar Organization and Magnetic Actuation of Flagella-like Nanoparticle Assemblies

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#### Videos of actuating 1H1T assemblies: Figure S1-S4

Video S1 corresponds to Figure 8 in the article. Video S2 is a similar video taken of another 1H1T assembly. Although both 1H1T assemblies appear to be propelling themselves through the DMF solvent, they may also be pulled towards the pole of the nearest electromagnet due to a magnetic field gradient. Without video showing two assemblies swimming in opposite directions under the same field at the same time, it is not possible to determine the cause of locomotion. That said, the 1H1T assemblies appear to swim in the direction of the head group along the y-axis of the magnetic field with a net velocity of approximately 1 µm/s.

<u>Video S3 is a control experiment showing 1H groups (PEG-MagNPs only) at a concentration of 10 µg/ml. Under identical magnetic field conditions as S1 and S2, the 1H groups do not appear to either rotate or translate in a particular direction. The undergo only random Brownian motion.</u>

<u>Video S4 is another control experiment showing pure 1T groups (PS-CoNPS only). In contrast</u> to the 1H groups, the 1T groups clearly rotate with the oscillation of the applied magnetic field. <u>However</u>, without a head group, the symmetrical reciprocal motion does not result in net locomotion. The 1T groups are slightly larger than the 1H groups, so the Brownian motion is not as pronounced. They do, however, undergo some Brownian motion. This motion does not result in net translation. This lack of translation contrasts with the 1H1T assemblies, which show a clear tendency to translate along the y-axis of the magnetic field.



Figure S3: Several frames of control experiment showing 1H groups undergoing random Brownian motion under the same field conditions as the video in Figure S1 and S2. Arrows point to 1H groups that are visible in the focal plane. The PEG-MagNP concentration was 10 µg/ml and the PS-CoNP concentration was 0. Conditions for the magnetic field:  $B_y = 0.15$  mT,  $B_x = 0.15$  mT RMS, f = 1.3 Hz.



Figure S4: Several video frames from a control experiment showing 1T groups with a PS-CoNP. The PS-CoNP concentration was 100  $\mu$ g/ml and the PEG-MagNP concentration was zero. Conditions for the magnetic field:  $B_v = 0.15$  mT,  $B_x = 0.15$  mT RMS, f = 1.3 Hz.

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## **Results of DOE screening study: Figure S5**

The absolute quantity of each assembly type is shown for each experiment. Note that the structures that are rich in head groups, such as 1H, >2H2T, and 3DG are represented in high numbers. This bias is partly due to the fact that PEG-MagNPs are easier to resolve in the optical microscope. We therefore normalized the data to the total number of observations for our statistical analysis.



### Prediction profile derived from the linear regression model: Figure <u>S6</u>

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The linear regression model predicts the fraction of each assembled species for a given set of conditions. Shown below is the prediction that maximizes the fraction of 1H1T assemblies. The red lines and the red values next to the axis labels indicate the exact values of each parameter. The white boxes indicate those parameters that had statistically significant correlations with the population distribution of assemblies.



#### **Results of the statistical t-test: Table S1**

The table below presents the data of Table II in more detail. "Prob>|t|" refers to the statistical significance of the correlation. 0.05 corresponds to a 95% confidence level. The t ratio indicates the strength of the correlation. A positive t ratio indicates direct proportionality and a negative t ratio indicates inverse proportionality. With respect to cross-correlations, a positive t ratio means that the two independent variables amplify the increase of a given species. A negative t ratio indicates that the fraction of a given species is highest when one dependent variable is large and the other is small. The sign of the t ratio also determines whether a quadratic correlation is U-shaped (positive) or I -shaped (negative).

Dependent Variable	Independent Variable	Prob> t	t Ratio
1 Head	PS-CoNP Conc.	0.0001	
	Magnetic Field Strength	0.0213	
	Time Before Sonication*Sonication Time	0.0053	
1 Tail	PEG-MagNP Conc.	0.0244	
1 Head 1 Tail	PS-CoNP Conc.	0.0126	
	Time Before Sonication*Sonication Time	0.0302	
2 Heads 1 Tail	PS-CoNP Conc.*PS-CoNP Conc.	0.0253	
	PEG-MagNP Conc.	0.0308	
	Time Before Sonication*Time Before Sonication	0.0471	
1 Head 2 Tails	PS-CoNP Conc.	0.0001	
	PS-CoNP Conc.*PS-CoNP Conc.	0.0224	
	Magnetic Field Strength	0.0074	
	Magnetic Field Strength*PS-CoNP Conc.	0.0001	
	Magnetic Field Strength*Time Before Sonication	0.0040	
	Time Before Sonication*Sonication Time	0.0007	
2 Heads 2 Tails	PS-CoNP Conc.	0.0044	
	PS-CoNP Conc.*PS-CoNP Conc.	0.0199	
	Sonication Time*Sonication Time	0.0320	
	Sonication Time*Time Before Sonication	0.0307	
	Sonication Time*Time After Sonication	0.0122	
3-D Globules	PS-CoNP Conc.	0.0160	
	PS-CoNP Conc.*PEG-MagNP Conc.	0.0374	
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