Supporting Information for: Self-propelling colloids with finite state dynamics
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Contents

1 SI 1: Microgel characterization: temperature-dependent size, electrophoretic mobility  2
2 SI 2: Estimation of the self-propulsion velocity of colloidal clusters  3
   A Microgel characterization via dielectric spectroscopy ........................................ 3
   B Theoretical estimation of EHDFs and dumbbell velocity ..................................... 3
3 SI 3: Temperature control with plasmonic heating: Experiments and FEM simulations  6
   A Illumination and light absorption characterization ........................................... 6
   B FEM modeling .................................................................................................. 6
4 SI 4: 3D-printed trap design for sequential capillarity-assisted particle assembly of microgels  9
5 SI 5: Yield of the sCAPA of R-G-PS trimers .......................................................... 10
6 SI 6: Estimation of the swimming velocity for R-G-PS trimers with different opening angles 11
7 SI 7: Determination of the chirality, rotational diffusion, and angular velocity of the active clusters 12
8 SI 8: Dynamical parameters of the experimental microswimmers .......................... 18
9 SI 9: Switching of chirality between states .......................................................... 21
10 SI 10: Calculation of microgel overlap on R-G-PS trimers ................................... 23
11 SI 11: Ensemble MSDs for chiral and non-chiral clusters .................................... 24
12 Supporting Movies ............................................................................................. 25
13 References ......................................................................................................... 26
1. SI 1: Microgel characterization: temperature-dependent size, electrophoretic mobility

The hydrodynamic size and electrophoretic mobility of the microgels were measured on a Malvern Instruments Zetasizer Nano ZS. Microgels do not have a well-defined charged surface; therefore, the electrophoretic mobility is reported instead of the $\zeta$-potential. The electrophoretic mobility experiments were performed in DTS1070 folded capillary cells with the microgels suspended in the same 0.1 mM HEPES buffer pH 7.4 used also in the AC-field driven active motion experiments. The hydrodynamic radius is measured in a quartz cuvette in 0.1 mM HEPES buffer pH 7.4. The temperature was controlled by the Zetasizer.

Fig. S1. A, Electrophoretic mobility of the G (squares) and R (triangles) microgels as a function of temperature. B, Hydrodynamic diameter of the G (squares) and R (triangles) microgels as a function of temperature. The error bars are the standard deviation of 5 measurements, which sometimes fall within the symbol.
2. SI 2: Estimation of the self-propulsion velocity of colloidal clusters

As mentioned in the main text, in order to estimate the propulsion velocity of the clusters in their different states, we need to have access to their temperature-dependent size (as reported above in Section SI 1) and temperature-dependent dielectric properties.

A. Microgel characterization via dielectric spectroscopy. The dielectric properties of the non-fluorescent PS particles and of the PNIPAM microgels were measured with a Novocontrol high-resolution dielectric analyzer (Alpha-A). The measurements were performed on a cell with a 6.45 mm gap distance between the electrodes enclosed by a Teflon cylinder, which was filled with the particle suspension at 0.1 wt%. The permittivity ($\epsilon'$), and conductivity ($\sigma'$) were determined over a wide frequency range (10²-10⁷ Hz) and varying the temperature from 20°C to 55°C with a rate of 2°C/min (Fig. S2). We further extracted $\epsilon'$ and $\sigma'$ corrected from the electrode polarization effect as a function of temperature, at the frequency of 800 Hz, i.e. the one used in the experiments to actuate the colloidal clusters (Fig. S3). We emphasize here that the dielectric spectroscopy measurements were carried out in fully de-ionized water, while the experiments are instead carried out in the presence of buffer and minute amounts of glucose remaining from the particle transfer. For this reason, the data reported below should only be considered appropriate to capture the trends in swimming velocity as a function of temperature and not as precise quantitative estimations. Nonetheless, we show that the predictions made from the measurements of dielectric properties in de-ionized water closely follow our experimental results, supporting our arguments. The dielectric spectroscopy measurements of a 0.1 % w.t sample of the PS particles used to fabricate the colloidal assemblies show only a minor dependence of $\epsilon'$ with temperature and a linear increase in $\sigma$ (Fig.S8), with no transition, as it is instead observed for the microgels.

B. Theoretical estimation of EHDFs and dumbbell velocity. As described in previous work (1), the measured dielectric properties can be used to calculate the magnitude and direction of the EHDF velocities $U_i$ (Fig.S4A,B), around a sphere under an AC electric field described as (2, 3)

$$U_i = \frac{\beta C K' + K'' \bar{\omega}}{\eta (1 + \bar{\omega}^2)^2} \frac{3(r_i/R_i)}{[1 + (r_i/R_i)^2]^{5/2}}$$

where $\bar{\omega} = \omega H/\kappa D$ with $\omega = 2\pi f$, $H$ twice the distance between the electrodes, $\kappa$ the Debye length, $D$ the diffusion coefficient of ions in solution and $\eta$ is the fluid viscosity. To evaluate the EHDF for every lobe of a dumbbell, we consider $r_i$ as the distance from the center of the evaluated lobe to the center of the adjacent particle ($r = R_{\text{gel}} + R_{\text{PS}}$). Therefore, as the radius of the microgel $R_{\text{gel}}$ varies with temperature, $r_i$ also changes. Finally, $K'$ and $K''$ are the real and imaginary part of the Clausius-Mosotti factor $K = K' + iK''$ (1, 4). To calculate the Clausius-Mosotti factor, we extract the particle’s $\epsilon_p$ and $\sigma_p'$ as described previously (1) from the corrected $\epsilon'$ and $\sigma'$ as a function of temperature and obtain them as

$$K' = \frac{\omega^2(\epsilon_p - \epsilon_m)(\epsilon_p + 2\epsilon_m) + (\sigma_p - \sigma_m)(\sigma_p + 2\sigma_m)}{\omega^2(\epsilon_p + 2\epsilon_m)^2 + 2(\sigma_p + 2\sigma_m)^2}$$

$$K'' = \frac{\omega(\epsilon_p - \epsilon_m)(\sigma_p + 2\sigma_m) - (\epsilon_p + 2\epsilon_m)(\sigma_p - \sigma_m)}{\omega^2(\epsilon_p + 2\epsilon_m)^2 + 2(\sigma_p + 2\sigma_m)^2}$$

where the subscript $m$ refers to the medium and $p$ refers to the particle.

Combining the $T$-dependent velocities of the EHDFs for each particle $U_i$, the dumbbell velocity $v_{\text{gel},PS}$ can, in first approximation, be obtained as a linear combination of the two values of $U_i$ (Fig.S4C) as

$$v_{\text{gel},PS} = \frac{U_{\text{gel}r_{PS}} + U_{PSr_{gel}}}{r_{gel} + r_{PS}}$$
Fig. S2. $\varepsilon'$ and $\sigma'$ as a function of frequency $f$ for an aqueous suspension at 0.1 wt % of red microgels (A-B), green microgels (C-D), and PS particles measured (E-F) for $T$ between 20 and 50°C.
**Fig. S3.** A, $\epsilon'$ and B, $\sigma'$ of the microgels (red and green solid symbols) and PS (gray open symbols) solution at 0.1 wt % (corrected from the electrode polarization effect) from Fig. S2 as a function of temperature $T$ for R (red circles) and G (green squares) microgels at a fixed frequency of 800 Hz.

**Fig. S4.** EHDF velocities $U_i$ calculated from Eq. (1) for A, Red (red solid line) and green (green solid line) microgels ($U_{\mu gel}$), and B, for the PS particle $U_{PS}$ close to each microgel (green and red dashed lines). The final $U_i$ value is obtained including the $\beta$ prefactor as one single free parameter $\beta_{PS} = 0.01 \beta_R = 0.075$ for the R-PS dumbbell and $\beta_{PS} = 0.07 \beta_G = 0.23$ for the G-PS dumbbell. C, Experimental values (symbols) and theoretical estimation (solid line) of dumbbell velocity as a function of temperature calculated from Eq. (4).
3. SI 3: Temperature control with plasmonic heating: Experiments and FEM simulations

A. Illumination and light absorption characterization. All experiments were performed on a NIKON Eclipse TI2-e microscope with a Lumencor Spectra II light source. To determine the intensity of the light leaving the microscope objective under different conditions, a Thorlabs S170C power sensor photo-diode in combination with a PM100D console was used. The sensor only measures the total light intensity and therefore was passed through a 0.0125 mm$^2$ pinhole to get the power density. Assuming a homogeneous illumination, the light intensity without the pinhole was used to calculate the area of the light spot. The results are summarized in Table S1. The transmittance of the Au layer was determined using the power density measured with an Au-coated slide between the objective and the sensor. The results are shown in Table S2. These illumination parameters are used in the Finite Element Method (FEM) model. Only the 20x objective with 395 nm and 555 nm illumination lines are used in experiments and only these parameters are modelled further. The light intensity for each wavelength can be controlled between 1 and 100% using the Nikon Imaging Software and these values are calibrated to give the corresponding power densities.

The heating due to light absorption of the Au layer was measured using a small thermo-couple connected to an OKO-lab temperature controller, which logs the temperature in 1 second intervals. The thermocouple was placed in the cell between the conductive Au-coated glass slides through the filling hole and the cell was filled with milli-Q water. The thermal gradient was measured by centering the objective first on the thermo-couple and subsequently moving it in controlled steps from this position with the motorized stage. The ambient temperature was logged with a separate thermo-couple and the heating was obtained as the difference between the temperature in the cell and the ambient temperature.

| Objective | Spot size (mm$^2$) | PD 640 nm (mW mm$^{-2}$) | PD 555 nm (mW mm$^{-2}$) | PD 395 nm (mW mm$^{-2}$) |
|-----------|------------------|-------------------------|-------------------------|-------------------------|
| 4x NA 0.13 | 13.73            | 3.2                     | 3.6                     | 2.9                     |
| 20x NA 0.45 | 0.55             | 48.0                    | 58.9                    | 45.0                    |
| 40x NA 0.6 | 0.16             | 89.1                    | 110.0                   | 86.2                    |
| 60x NA 0.7 | 0.07             | 109.2                   | 139.9                   | 107.9                   |

Table S1. Power densities and spot size for different objectives

| Wavelength (nm) | Transmission (%) | Absorption (%) | Reflection (%) |
|-----------------|------------------|----------------|----------------|
| 395             | 27               | 55             | 18             |
| 555             | 38               | 41             | 21             |
| 640             | 37               | 35             | 28             |

Table S2. Transmission, Absorption and Reflection of the 10 nm Au-layer

B. FEM modeling. The FEM model is constructed in COMSOL Multiphysics (5) to be an accurate description of our setup, but simplifications are made to ease computation. A schematic overview is shown in Figure S5. The model is a time-dependent FEM 2D simulation with axial symmetry and uses the in-build "Heat transfer in solids and Fluids (ht)", "Laminar flow (spf)" and "Nonisothermal Flow (nitf1)" physics simulations from COMSOL. Therefore, both the diffusive and convective heat transfers are modelled. The plasmonic heating in the thin gold films is not directly modeled, but we accounted for it in a simplified way by including heat sources over an area corresponding to the light spot on both the top and bottom slides. The magnitude of the heat source on the bottom is determined by the power density of the light and the expected absorption of the Au layer (the values for 395 nm are used unless stated otherwise). The heat source on the top is determined from the power density that is transmitted through the bottom Au layer taking both absorption and reflection into account. The top and bottom slides are expected to be cooled via
convection. The edge of the bottom slide is in direct contact with the metal of the microscope stage which acts as a heat sink and is modelled as a constant temperature. The model is compared to values measured with a temperature probe and the sensitivity to mesh size and time step is also characterized.

The FEM model has an excellent agreement with experiments in both the power scaling (Figure S6A) and the thermal gradients (Figure S6B). There is a discrepancy in the initial time-dependent heating (Figure S6B), however, this is likely a limitation of the experimental method due to the non-negligible thermal mass of the thermal probe.

Fig. S5. Schematic description of the COMSOL model. The dimensions are not in scale.

Fig. S6. Comparison between experiments and simulations. A, Heating as a function of light intensity, measured with the thermo-couple (squares) and modelled with COMSOL (triangles), in the center of the illuminated spot after 60 s for the 395 nm line. B, Thermal profile measured from the center of the illuminated spot (r = 0) in experiments (circles) and simulations (solid line) after 60 s at a power density of 45 mW mm\(^{-2}\). C, Time-dependent heating measured experimentally (circles) and modelled (solid line) at the center of the illuminated spot for a power density of 45 mW mm\(^{-2}\).
Fig. S7. Sensitivity analysis of the COMSOL model for modelling parameters. A, Analysis of the sensitivity as a function of the time step for the heating in the center at 45 mW mm\(^{-2}\) and \(t = 60\) s. B, Analysis of the sensitivity as a function of the mesh size for the final heating in the center at 45 mW mm\(^{-2}\) and \(t = 60\) s. The text above the data points is the mesh settings used in COMSOL. The dashed lines mark the experimentally measured heating.
Fig. S8. A, Side and top view of the 3D-model for the sCAPA-masters for the 120-cluster. The dimensions for the different parts are given in µm. B, Side and top view of the 3D-model for the sCAPA-masters for the 60-cluster. C, Side and top view of the 3D-model for the sCAPA-masters for the 180-cluster. D, SEM-micrograph of the 3D-printed sCAPA-masters for the 120-cluster. E, SEM-micrograph of the 3D sCAPA-traps for the 120-cluster. F, SEM-micrograph of the 3D sCAPA-traps for the 120-cluster filled with the PS-particles. Scale bars are 5 µm
5. SI 5: Yield of the sCAPA of R-G-PS trimers

Fig. S9. Yields of the sCAPA for A, 60 cluster B, 120-clusters, and C, 180-clusters. The inset images are representative of the most common errors for the different depositions. These errors are a surplus of green-microgels in the wrong places (green-rich) or a lack of red or green microgels (red-poor or green-poor).
6. SI 6: Estimation of the swimming velocity for R-G-PS trimers with different opening angles

We use a simple triangular vector model to estimate the final swimming velocity $v$ for a trimer with internal angle $\theta$. We decompose the propulsion of the trimer into the one generated by each PS-microgel dumbbell and consider their linear superposition (Fig.S10A). In particular, we obtain the two in-plane velocity components for each dumbbell as

$$v_{x,i} = v(T) \frac{\cos \theta}{2};$$  \hspace{1cm} [5]

$$v_{y,i} = v(T) \frac{\sin \theta}{2};$$  \hspace{1cm} [6]

where $v(T)$ is the velocity magnitude as a function of temperature for each dumbbell, obtained from Fig.S4C for the experimental and theoretical data, $\theta$ the opening angle between the microgels attached to a PS particle, and $\hat{x}$ and $\hat{y}$ are the unit vectors in x and y, respectively. Finally, we sum the $v_{x,i}$ and $v_{y,i}$ components to obtain the trimer velocity $v$ as:

$$v = [(v_{x,R} + v_{x,G}), (v_{y,r} + v_{y,g})].$$  \hspace{1cm} [7]

We finally calculate the magnitude of $v$ as a function of the temperature $T$ for each case as $\|v\| = \sqrt{(v_x)^2 + (v_y)^2}$ (Fig.S10B).

![Fig. S10. A, Scheme of a R-G-PS trimer, with an opening angle $\theta$ between the microgels. The green and red arrows represent the velocity vectors for each PS$_{dumb}$ pair, obtained from the data fitted in Fig.S4. The black arrow indicates the sum velocity vector. B, Estimated trimer velocities $v$ as a function of temperature for each experimental configuration with $\theta$ 60 °, 120 °, and 180 ° (black, red, and blue respectively). The symbols indicate the result using the experimental dumbbell velocity, and the solid lines are fits the data, using the vectorial sum of the expressions obtained from Eq. 4 for each dumbbell. The large open symbol represents the experimental velocities of the different clusters.](image)
7. SI 7: Determination of the chirality, rotational diffusion, and angular velocity of the active clusters

To establish whether a trajectory can be considered statistically chiral or not, one first needs to consider that non-chiral active Brownian particles can appear chiral if observed over timescales short than or similar to the rotational diffusion time \( \tau_r = 1/D_r \). Therefore, in order to classify trajectories one needs to carefully examine the experimental distribution of angular displacements and establish a comparison with the corresponding distribution of an active Brownian particle with no chirality.

To this end, we look at the distribution of angular displacements (\( \Delta \phi \)) of a trajectory at different lag times (\( \delta t \)). (Figure S11). At each time step, the direction of motion is determined from the ratio of the \( x \) and \( y \) displacements as \( \phi = \arctan(\Delta y/\Delta x) \), with \( \Delta x = x_{t+1} - x_t \) and \( \Delta y = y_{t+1} - y_t \). Then the angular displacements are given by \( \Delta \phi = \phi_{t+\delta t} - \phi_t \). The mean cumulative angular displacements for a chiral swimmer scale with \( \Delta \phi = \omega \delta t \), while the standard deviation of that distribution is given by \( \sigma \Delta \phi = 2 \sqrt{D_r \delta t} \). Conversely, a non-chiral ABP has zero mean cumulative angular displacements but the same standard deviation.

To validate our analysis and estimate the relative errors, we simulate active Brownian particles with chiral motion and extract their angular displacements as mentioned above. The model we use is:

\[
\dot{x}(t) = v \cos(\phi(t)) + \sqrt{2D_t} \xi,
\]

\[
\dot{y}(t) = v \sin(\phi(t)) + \sqrt{2D_t} \xi,
\]

\[
\dot{\phi}(t) = \omega + \sqrt{2D_r} \xi,
\]

With \( v \) the propulsion velocity, \( D_t \) the translational diffusion, \( D_r \) the rotational diffusion, \( \omega \) the angular velocity, and \( \xi \) a Gaussian white noise term. We simulate 100 particles for 30 s with a time step of 0.1 s (Figure S11). These values are comparable with the experimental conditions. Then we extract the mean and standard deviation of the angular displacement distributions at multiple lag times and fit the resulting curves with a simple linear regression (Figure S12). The advantage of using the mean angular displacements as opposed to the mean squared angular displacements is that the sign of the angular velocities, i.e. clockwise or counterclockwise rotation, can be determined. The number of data points scales with \( n_{\delta t} = n - \delta t \), thus lag times close to total time suffer from poor statistics and are discarded.

From the plots in Figure S11 the requirement to distinguish chiral from non-chiral active Brownian particles becomes clear; if the mean \( \langle \Delta \phi \rangle \) evaluated at \( \delta t \) falls outside the standard deviation \( \sigma \langle \Delta \phi \rangle \) there is a 67% chance that the particle has a non-zero angular velocity. Therefore, the resulting criterion for chirality is \( \omega \delta t > 2 \sqrt{D_r \delta t} \). As one can expect, evaluating at longer times allows the verification of smaller angular velocities or noisier trajectories.

In Figure S13, we show the results of a sensitivity analysis of this method to different parameters. We observe that the error in \( \omega \) increases roughly linearly with the rotational diffusion (Fig. S13A), while \( D_r \) stays constant. Moreover, errors are larger for lower angular velocities (Fig. S13B) as there is a smaller difference between \( \omega \) and \( D_r \). Furthermore, the range of lag times is important for accuracy. Using long lag times reduces the statistics in the longer time scales and therefore gives less accurate results (Fig. S13C). Furthermore, the sampling is staggered thus there are some unintended correlations that are especially prominent in the long lag time regime. Comparing these results with the values reported in the main text and in Section S1 8, we note that errors in the estimation of \( \omega \) are typically below 20%, and most often below 10%. This approach works in the same way for the experimental data (Figures S14 and S15). Here, each portion of the trajectories corresponding to the different states is 30 s long and is evaluated for a max \( \delta t \) of 5 s with 20 time steps (only 5 are shown in Figures S14 and S15). Additionally, we also need to account for the spread in the angular displacements coming from the accuracy of particle tracking. However, the contribution of this additional noise is independent of time and will just result in a constant shift. The results for all trajectories for both \( D_r \) and \( \omega \) are shown in Section S1 8. The mean values are used to compute the MSD of the “average particle”, which results in the MSD curves shown in the main manuscript (Figure 5D), computed as following the expression of
the MSD of a chiral swimmer derived by Archer et al. (6):

\[ MSD(t) = 4D_t t + \frac{2v^2 + D_r t}{D_r^2 + \omega^2} + \frac{2v^2(\omega^2 - D_r^2)}{(D_r^2 + \omega^2)^2} + \frac{2v^2e^{-D_r t}}{(D_r^2 + \omega^2)^2} \left[ (D_r^2 - \omega^2) \cos \omega t - 2\omega D_r \sin \omega t \right] \]

Fig. S11. Left: Distributions of angular displacements \( P(\Delta \phi) \) for \( \delta t = 0.1, 1, 2, 3, 4, 5 \) s for simulated chiral active Brownian particles with, \( D_r = 0.5, \ \text{rad}^2 s^{-1}, \ \omega = 0 \ \text{rad} s^{-1} \) (A, black), \( D_r = 0.5, \ \text{rad}^2 s^{-1}, \ \omega = 1 \ \text{rad} s^{-1} \) (B, red), and, \( D_r = 0.8, \ \text{rad}^2 s^{-1}, \ \omega = -1.5 \ \text{rad} s^{-1} \). Right: the corresponding trajectories from which the distributions are extracted.
Fig. S12. Evolution of the mean angular displacements (A) and their variance (B) as a function of lag time. The symbols correspond to trajectories shown in Figure S11 A (crosses), B (squares), and C (circles) with the corresponding $D_r$ and $\omega$. The lines in A are the predicted mean of the distributions and the shaded area is the standard deviation for the corresponding $D_r$ and $\omega$. The lines in B are the predicted variance for the corresponding $D_r$. 
Fig. S13. Sensitivity analysis to determine $D_r$ and $\omega$ using simulated trajectories of 30 s. The error is the average relative deviation from the input values of the simulations ($\text{error}_\omega = |\omega_{\text{cal}} - \omega_{\text{sim}}| / \omega_{\text{sim}}$) as a function of $D_r$, $\omega$, and $\delta t_{\text{max}}$. In each of the parameter sweeps, one parameter is varied and the other two are fixed. Left column) The error in $\omega$ (A) and $D_r$ (D) for increasing $\omega$ with a fixed $D_r = 0.5$, $\delta t_{\text{max}} = 5$. Middle column) The error in $\omega$ (B) and $D_r$ (E) for increasing $D_r$ with a fixed $\omega = 1.5$, $\delta t_{\text{max}} = 5$. Right column) The error in $\omega$ (C) and $D_r$ (F) for increasing $\delta t_{\text{max}}$ with a fixed $D_r = 0.5$, $\omega = 1.5$. 
Fig. S14. Example analysis for one trajectory in state (GR), (G'R), and (G'R') for the median 120-cluster, showing the distribution of angular displacements at $\delta t = 0.1, 1, 2, 3, 4, 5$ s and the corresponding trajectory for state (GR) (A), (G'R) (B), and (G'R') (C).
Fig. S15. The evolution of the mean angular displacements (A) and their variance (B) as a function of the lag time for the different states (GR) (circles), (GR') (crosses), and (GR') of a median 120-cluster, for the trajectories and angular displacement distributions shown in Figure S14. The lines are the corresponding least square fits.
8. SI 8: Dynamical parameters of the experimental microswimmers

In the tables below we show the mean and standard deviation of the angular velocities, self-propulsion velocities, and rotational diffusivities for each cluster type (60-clusters: Table S3, 120-clusters: Table S4, and 180-clusters: Table S5). These values are extracted from the data using the distribution of angular displacements as described in Section SI 7. We further report three experimental trajectories with parameters that most closely represent the median values of the angular velocities, self-propulsion velocities, and rotational diffusion for each cluster type (Figure S16). They qualitatively exemplify the types of motion most common in the different states, supporting the finding discussed in the main text. Furthermore, we also report the values of rotational diffusion (Figure S17) and angular velocity for all trajectories in box plots (Figure S18). From the former, we observe that the trimers show a strong increase in $D_r$ in the different states; already in $(G'R)$ for the 60-clusters, and then in $(G'R')$ for the 120- and 180-clusters. Upon returning to state $(G'R)$, the distributions of $D_r$ recover to the initial values.

The angular velocities show less pronounced differences across the different states for all clusters. The data that we report correspond only to those trajectories that are statistically chiral, based on the arguments presented above ($\langle |\omega| \rangle t > 2 \sqrt{D_r t}$ for $t = 30$ s). Therefore for particles with a higher $D_r$, a higher $\omega$ is needed to reach that threshold. Therefore, for example, for state $(G'R')$, the angular velocities of the 120- and 180-clusters seem to increase, but that is because only the fastest rotating clusters can be statistically discerned from non-chiral trajectories. Consequently, the fraction of chiral swimmers as shown in Figure 5B is a better descriptor of the variations of particle dynamics across the various states as opposed to the absolute values of $\omega$.

| 60 -clusters | GR | G'R | G'R' |
|--------------|----|-----|------|
| $N_{\text{trajectories}}$ | 52 | 79  | 67   |
| $\langle |\omega| \rangle$ (rad s$^{-1}$) | 0.31 | 0.28 | 0.18 |
| $\sigma |\omega|$ (rad s$^{-1}$) | 0.65 | 0.47 | 0.37 |
| $\langle v \rangle$ (µm s$^{-1}$) | 4.96 | 1.74 | 2.09 |
| $\sigma v$ (µm s$^{-1}$) | 1.18 | 0.66 | 0.61 |
| $\langle D_r \rangle$ (rad$^2$ s$^{-1}$) | 0.48 | 2.47 | 2.73 |
| $\sigma D_r$ (rad$^2$ s$^{-1}$) | 0.55 | 1.00 | 0.95 |

Table S3. Dynamical parameters of the 60-clusters

| 120 -clusters | GR | G'R | G'R' |
|---------------|----|-----|------|
| $N_{\text{trajectories}}$ | 105 | 106 | 104 |
| $\langle |\omega| \rangle$ (rad s$^{-1}$) | 0.40 | 0.51 | 0.24 |
| $\sigma |\omega|$ (rad s$^{-1}$) | 0.46 | 0.49 | 0.43 |
| $\langle v \rangle$ (µm s$^{-1}$) | 3.42 | 4.72 | 2.55 |
| $\sigma v$ (µm s$^{-1}$) | 0.56 | 0.82 | 0.53 |
| $\langle D_r \rangle$ (rad$^2$ s$^{-1}$) | 0.33 | 0.58 | 2.18 |
| $\sigma D_r$ (rad$^2$ s$^{-1}$) | 0.47 | 0.43 | 0.94 |

Table S4. Dynamical parameters of the 120-clusters
Table S5. Dynamical parameters of the 180-clusters

| 180 -clusters | GR | G'R | G'R' |
|---------------|----|-----|------|
| $N_{\text{trajectories}}$ | 78 | 103 | 144 |
| $\langle |\omega| \rangle$ (rad s$^{-1}$) | 0.23 | 0.22 | 0.37 |
| $\sigma |\omega|$ (rad s$^{-1}$) | 0.38 | 0.49 | 0.47 |
| $\langle v \rangle$ ($\mu$m s$^{-1}$) | 2.16 | 3.76 | 2.36 |
| $\sigma v$ ($\mu$m s$^{-1}$) | 0.47 | 1.10 | 0.53 |
| $\langle D_r \rangle$ (rad$^2$ s$^{-1}$) | 0.52 | 0.62 | 2.34 |
| $\sigma D_r$ (rad$^2$ s$^{-1}$) | 0.69 | 0.41 | 0.98 |

Fig. S16. Example trajectories for the 60-clusters (A), 120-clusters (B), and 180-clusters (C). The trajectories are color-coded by temperature according to the heating sequence shown in Figure 4B. These trajectories have a self-propulsion velocity, angular velocity, and rotational diffusion that most closely represent the ensemble average of the corresponding populations. Scale bars are 20 µm

Fig. S17. Box plot of the mean rotational diffusion in $GR$, $G'R$, or $G'R'$ for the illumination sequence described in Figure 4B. The box plots are calculated from between 52 to 79, 106 to 104, and 78 to 144 trajectories for the 60-, 120- or 180-clusters, respectively. The box indicates quartiles, the colored line the median, the bars the whiskers (Q+1.5*IQR), and the dots the outliers.
Fig. S18. Box plot of the mean absolute angular velocity in $GR$, $G'R$, or $G'R'$ for the illumination sequence described in Figure 4B. The box plots are calculated from between 3 to 25, 11 to 83, and 21 to 54 trajectories for the 60-, 120- or 180-clusters, respectively. Here, only the chiral trajectories are considered for the extraction of the angular velocity, and the corresponding fraction of chiral trajectories is shown in Figure 5B. The box indicates quartiles, the colored line the median, the bars the whiskers (Q+1.5*IQR), and the dots the outliers.
9. SI 9: Switching of chirality between states

In our experiments, we sometimes observe switching between chiral and non-chiral motion when transitioning between GR and G’R, as well as flipping between clockwise and counterclockwise chirality between GR at the beginning and the end of the illumination sequence. These events are characterized and quantified below.

The switch between chiral and non-chiral motion when transitioning between GR and G’R is most evident in the 120-clusters where the fraction of chiral swimmers is highest (Figure S19). Although in Figure 5B the fraction of chiral swimmers is constant, the details are more nuanced. About 20% of the swimmers exhibit a switch, where roughly equal fractions switch from chiral to non-chiral motion and vice versa, thus the total fraction of chiral swimmers remains similar. The exact mechanisms for the different behavior most likely vary from cluster to cluster but we hypothesize that can be generally explained in relation to the different trajectory types already present in GR. If a cluster has a reasonably balanced propulsion generated by both microgels, one would observe a non-chiral trajectory, that may become chiral if the G microgel collapses (Figure S19B). Conversely, an asymmetrically propelled cluster with a stronger EHDF generated by G than R in state GR may become more symmetric and non-chiral when the G microgel collapses (Figure S19C).

Although most swimmers recover chiral motion between the initial and final state GR, we observed that it is not uncommon for the direction of chirality to be in the opposite direction at the end of the illumination sequence. We characterize the fraction of trajectories that flip chirality in Figure (S20). We see that around 25% of the trajectories flip chirality and this is observed consistently between the different cluster types. We hypothesize that the clusters in state G’R’ are not only experiencing enhanced rotational diffusion in the plane but are also free to rotate out of the plane.

Fig. S19. A) The left bar shows the fraction trajectories for the 120-cluster that switches from chiral to non-chiral motion. The right bar shows, for those particles that switch, which ones switch from non-chiral motion in GR to chiral motion in G’R (GR → G’R'), or from chiral motion in GR to a non-chiral motion in G’R (GR ← G’R'). B) An example of a swimmer switching from non-chiral to chiral. C) An example of a swimmer switching from chiral to non-chiral. Scale bars are 20 µm.
Fig. S20. Fraction of trajectories that flip chirality in state GR between the beginning and the end of the illumination sequence. A, B and C correspond to the 60-, 120-, 180-clusters, respectively. The plots are obtained from between 74 to 48, 106 to 37, and 79 to 26 trajectories for the 60-, 120- or 180-clusters, respectively. The inset in panel B is an example of a trajectory whose chirality flips from clockwise to counterclockwise rotation. Scale bar is 20 µm.
10. SI 10: Calculation of microgel overlap on R-G-PS trimers

Here, we report simple geometrical calculations to estimate the overlap of the two microgels attached to the PS particle at different opening angles \( \theta \). Based on the scheme in Figure S21A, we obtain the following relationships:

\[
\begin{align*}
 h_R &= h_G = h \\
 d &= 2r_{PS} \times \sin \left( \frac{1}{2} \theta \right) + 2h \times \sin \left( \frac{1}{2} \theta \right)
\end{align*}
\]  

Here, \( r_{PS} \) is the radius of the hard PS colloid (1 \( \mu \)m), \( r_R \) is the radius of the red-core microgel, \( r_G \) is the radius of the green-core microgel and \( h_R \) and \( h_G \) are the distances at which the microgels’ centers sit from the PS surface. The radii are easily found with the DLS data (Figure ??B) and are 1.15 \( \mu \)m and 1.3 \( \mu \)m for the G and R microgels, respectively. However, the height from the surface is more difficult to determine. Here we use atomic force microscopy (AFM) in water to probe the configuration of the microgel on the particle surface (Figure 10B). The AFM does not probe the full microgel as the sharp tip cannot easily detect the loosely cross-linked polymer chains in the corona at the periphery of the particle and only the more-densely cross-linked core is clearly visible. However, AFM images can still be used to measure the distance \( h \). We assume that \( h_R = h_G = h \) measures a value of approximately 400 nm.

From these simple geometrical considerations, we find that our microgels start to overlap at an opening angle \( \theta \leq 120^\circ \). This confirms that there is significant overlap for the 60-clusters, while no overlap is found for the 120- and 180-clusters. These results indicate some simple design guidelines to achieve multiple states for particles and microgels of different sizes.

Fig. S21. Calculation of the theoretical overlap between the microgels adsorbed on a PS particle. A, Schematic of the geometry of the problem used for the calculation. B, AFM micrograph of an R-PS dumbbell in water. Scale bar is 1 \( \mu \)m.
11. SI 11: Ensemble MSDs for chiral and non-chiral clusters

In the main manuscript, we treat the populations of microswimmers as a whole, however, we have a mixture of chiral and non-chiral swimmers in our experiments. Here, we show the ensemble-average MSDs of the chiral and non-chiral separately to acknowledge the fundamental differences between these two modes of motion. Where a non-chiral ABP is characterized by ballistic behavior at short timescales \( (t < \tau_r) \) and enhanced diffusive motion at long timescales \( (t > \tau_r) \), chiral swimmers loop back to the same spot. The latter fact results in MSDs with a characteristic sinusoidal shape with a period of \( 2\pi \omega \). However, the presence of multiple microswimmers with a distribution of angular velocities washes out the regular oscillations and leads to the appearance of a shoulder in the MSD as shown in the figures below.

![Fig. S22. Ensemble-average mean-squared displacements of the chiral (red dotted lines) and non-chiral ABP (blue solid lines) fractions shown in Figure 5B. The columns correspond to the 60-, 120-, and 180-clusters. The rows correspond to the different states GR, G'R, and G'R'. The shaded areas correspond to the 95% confidence intervals. The errors are calculated from between 52 to 79, 106 to 104, and 78 to 144 trajectories for the 60-, 120- or 180-clusters, respectively.](image-url)
12. Supporting Movies

- **Movie S1**: Movie of the active motion of a 120-cluster under an AC field (800Hz, 7V Vpp) recorded at 10 fps and sped up three times for visualization. The particle trajectory is color-coded by the temperature, as described in the main text. Only the PS particle is visible in the bright-field images.

- **Movie S2**: Movie of the active motion of a 60-cluster under an AC field (800Hz, 7V Vpp) recorded at 10 fps and sped up three times for visualization. The particle trajectory is color-coded by the temperature, as described in the main text. Only the PS particle is visible in the bright-field images.

- **Movie S3**: Movie of the active motion of a 180-cluster under an AC field (800Hz, 7V Vpp) recorded at 10 fps and sped up three times for visualization. The particle trajectory is color-coded by the temperature, as described in the main text. Only the PS particle is visible in the bright-field images.
13. References

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