Supporting Information

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SI Text

The geometry of a chiral tetramer is shown in Fig. S6. Three petal dimers are labeled in red, blue, and green, respectively. The pink line indicates the long axis of one petal. The torque on a chiral tetramer can be expressed as

$$T = NF_d L_a, \qquad \text{[S1]}$$

where N is the number of petals, F_d is the drag force on one petal, and $L_a = AB$ is the arm length. L_a can be found geometrically through the relation

$$L_a = AE \cdot \cos\left(\frac{\pi}{2} - \frac{\pi}{3} - \alpha\right), \qquad \text{[S2]}$$

where $AE = R_2/\sin(\pi/3)$ and $\alpha = \arcsin[R_2/(R_1 + R_2)]$.

The drag force on a propelling lying dimer is approximately

$$F_d = 6\pi\mu(R_1 + R_2)U, \qquad \text{[S3]}$$

where U is the propelling speed of an individual dimer. The angular velocity of a chiral cluster Ω_0 can be related to torque by

$$T = 8\pi\mu R^3 \Omega_0, \qquad \text{[S4]}$$

where R is an effective radius of the chiral cluster if we approximate it as a sphere. Combining Eqs. S1–S4, we can find the ratio between Ω_0 and U:

$$\frac{\Omega_0}{U} = \frac{3N(R_1 + R_2)L_a}{4R^3}. \qquad \text{[S5]}$$

With $R_1 = 1.27$ μm, $R_2 = 0.89$ μm, and $R \sim 2R_1$, Eq. **S4** provides the slope of the solid line shown in Fig. 4D.
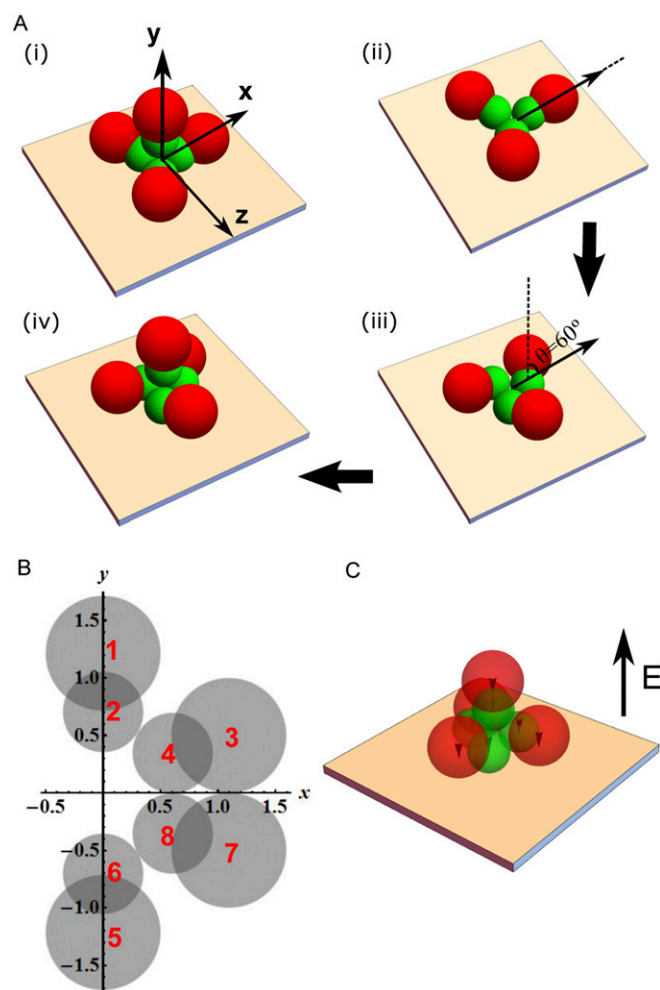
Fig. S1. (*A*) Coordinates of a 3D tetramer used in computation. The substrate corresponds to the $x-z$ plane. (*A, i* and *ii*) Achiral cluster where the long axis of one lying dimer petal is aligned with the x axis. Any nonzero angle between the dimer's long axis and the x axis results in a chiral cluster, as exemplified in *A, iii* with a rotation angle of 60°. (*B*) A lying dimer petal (spheres 3 and 4) and a central dimer (spheres 1 and 2) projected in the $x-y$ plane in an achiral tetramer. Dimers located in the bottom half plane (spheres 5–8) are image dipoles that account for surface conductive boundary conditions. Replicating the petal with 120° rotations about the y axis generates the tetramers shown in *A, i*, where image dipoles beneath the substrate are not shown for clarity. (*C*) The calculated dipoles for each constitutive dimer in a chiral tetramer. They point to a direction that is antiparallel to the applied field because of their negative polarizabilities.
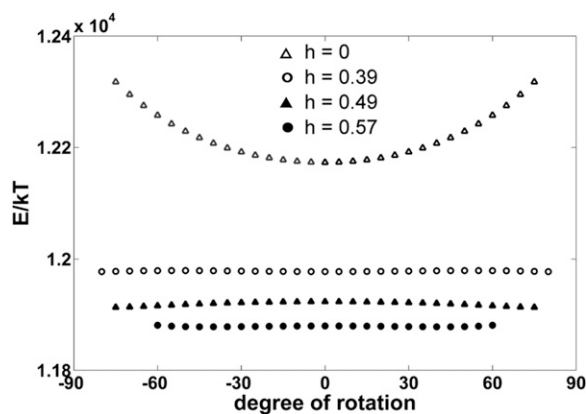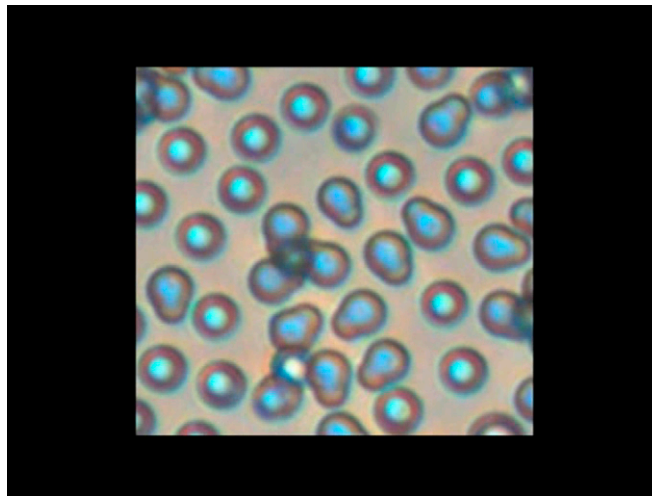

Fig. S2. The electrostatic energy of a tetramer built by four dimers with different separation h between the central dimer and the substrate and petal rotation angle θ. h has been scaled by the diameter of the large lobe $2R_1$. The building block (i.e., the dimer) has lobe sizes of $R_1 = 1.27$ μm and $R_2 = 0.89$ μm. The bond length L is 1.25 μm. $V_{pp} = 20$ V is applied across 100 μm.

Fig. S3. The electrostatic energy of clusters with increasing number of petals, ranging from two (trimer) to five (hexamer). *h* is chosen to be the maximum so that each curve represents the global minimum.

Fig. S4. The impact of electric field strength on the stability of chiral tetramers.

Fig. S5. The impact of asymmetry in polarization coefficient on the formation of chiral tetramers from geometrically symmetric dimers. The dimer has lobe sizes of $R_1 = R_2 = 1.27$ μm. The bond length *L* is 1.52 μm. $V_{pp} = 10$ V is applied across 100 μm. The polarization coefficient of lobe 1, $K_1 = -0.5$, whereas K_2 varies from -0.5 to 0.5.

Fig. S6. Schematics illustrate the calculation of arm length L_a.

Fig. S7. The impacts of electric field strength and frequency on the angular velocity of a chiral tetramer. (*A*) The angular velocity is proportional to the square of the field strength at 1,000 Hz, consistent with an induced EHD flow propulsion mechanism. (*B*) The angular velocity decreases with increasing frequency.

Movie S1. The assembly of chiral and achiral colloidal clusters induced by AC electric fields. The asymmetric dimer has lobe sizes of $R_1 = 1.27$ μm and $R_2 = 0.89$ μm. The bond length is 1.25 μm. Part 1: Chiral cluster formation. At 1 V_{pp} (peak-to-peak voltage) and 700 Hz, the dimers move randomly. When voltage is increased to 10 V_{pp} at 700 Hz, some dimers will align with the external field (i.e., stand on the substrate), while others remain lying on the substrate. The standing dimer attracts one lying dimer and together they form a pair. Two more lying dimers join in the assembly one by one. The petals then rearrange into a chiral configuration. Part 2: Achiral cluster formation. At 1 V_{pp} and 1,000 Hz, the dimers move randomly. When voltage is increased to 12 V_{pp} at 1,000 Hz, some dimers will align with the external field (i.e., stand on the substrate), while others remain lying on the substrate. However, the standing dimers can adopt two different orientations: smaller lobe pointing toward the substrate or away from the substrate. When lying dimers associate with a standing dimer with its large lobe pointing toward the substrate, achiral clusters can be formed. Part 3: The chiral cluster can be permanently fixed on the substrate by combining an AC electric field (600 Hz, 13 Vpp) and DC pulses (3.2 V for 0.5 s). The electrophoretic force drives all petals toward the substrate and they are fixed on the substrate due to strong van der Waals forces. The chirality is maintained even after both AC and DC electric fields are turned off. Note that the central standing dimer cannot be fixed. When the electric field is turned off, it undergoes Brownian motion and eventually leaves the cluster. All movies are displayed in real time.
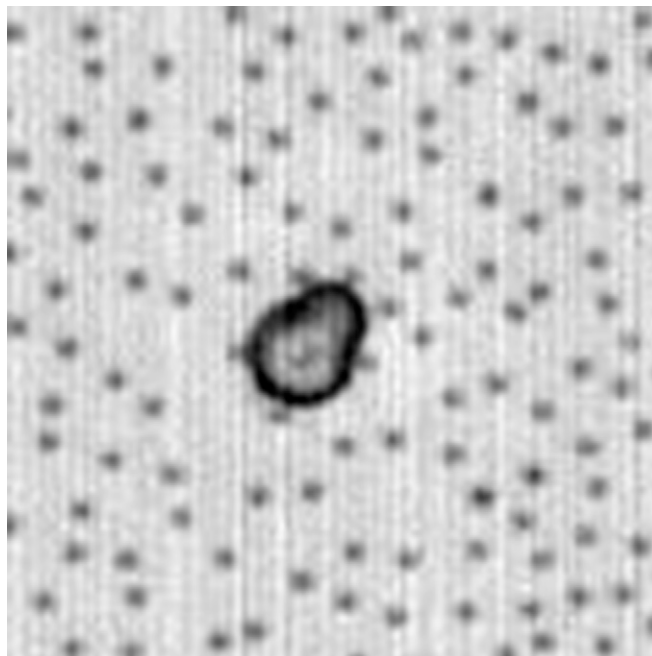
[Movie S1](#)
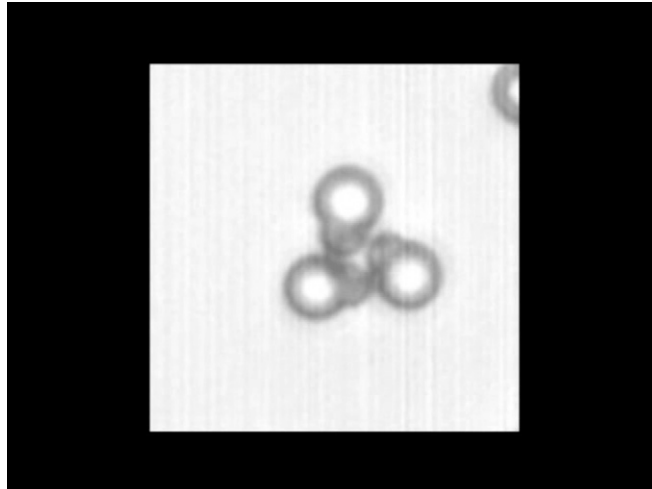
Movie S2. The rotation of chiral clusters under AC electric fields. The asymmetric dimer has lobe sizes of $R_1 = 1.35$ μm and $R_2 = 0.91$ μm. The bond length L is 1.43 μm. The right-handed chiral tetramer rotates counterclockwise, and the left-handed chiral tetramer rotates clockwise. As a comparison, the achiral heptamer stays stationary. Experimental information: applied voltage $V_{pp} = 13$ V; frequency, 500 Hz; gap between two electrodes, 100 μm. The movie is displayed in real time.
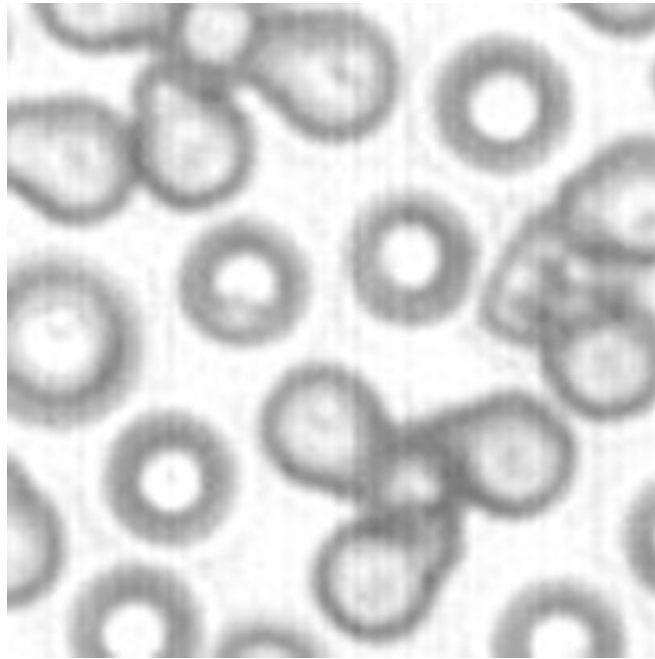
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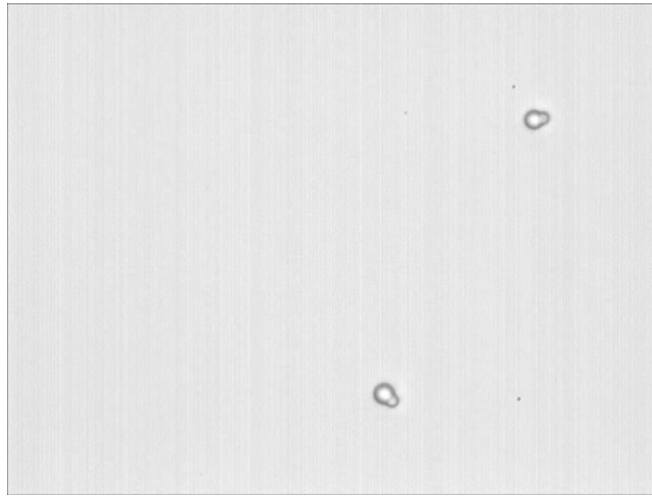
Movie S3. The linear propulsion of asymmetric dimers under AC electric fields. The asymmetric dimer has lobe sizes of $R_1 = 1.35$ μm and $R_2 = 0.91$ μm. The bond length L is 1.43 μm. Part 1: The standing dimer does not propel under electric field due to a balanced electrohydrodynamic flow surrounding the particle. Part 2: An asymmetric lying dimer, however, moves with its smaller lobe facing forward. Experimental information: applied voltage $V_{pp} = 10$ V; frequency, 800 Hz; gap between two electrodes, 100 μm. The movie is displayed in real time.

[Movie S3](#)

Movie S4. The unbalanced electrohydrodynamic flow surrounding an asymmetric lying dimer under AC electric fields. The asymmetric dimer has lobe sizes of $R_1 = 1.27$ μm and $R_2 = 0.89$ μm. The bond length L is 1.25 μm. In this movie, we purposely immobilize the dimer on an ITO substrate (due to van der Waals attraction). The movement of tracer particles (500 nm in diameter) then indicates the unbalanced electrohydrodynamic flow surrounding the dimer: The flow is directed toward the smaller lobe but moves away from the larger lobe. When the dimer is freely suspended, it moves with its smaller lobe facing forward. Experimental information: applied voltage $V_{pp} = 10$ V; frequency, 800 Hz; gap between two electrodes, 100 μm. The movie is displayed in real time.

[Movie S4](#)

Movie S5. Tuning the chirality of a tetrameric cluster. In this movie, we present a method to tune the chirality of a tetrameric cluster from right handed to left handed. Initially, the right-handed cluster is stable at 600 Hz. When we increase the frequency to 1,800 Hz, it becomes unstable and the chirality fluctuates due to Brownian motion. At the moment when the chirality switches to left handed, we quickly decrease the frequency back to 600 Hz so that its handedness can be locked. Although it is done manually, the tuning can be performed, in principle, with automation based on image processing. By combining our method with microfluidics, one can possibly make chiral clusters one by one with fully controllable handedness.

Movie S5