

Supplementary Material for

Geometric programming of asymmetric colloidal dynamics via topological defect reconfiguration

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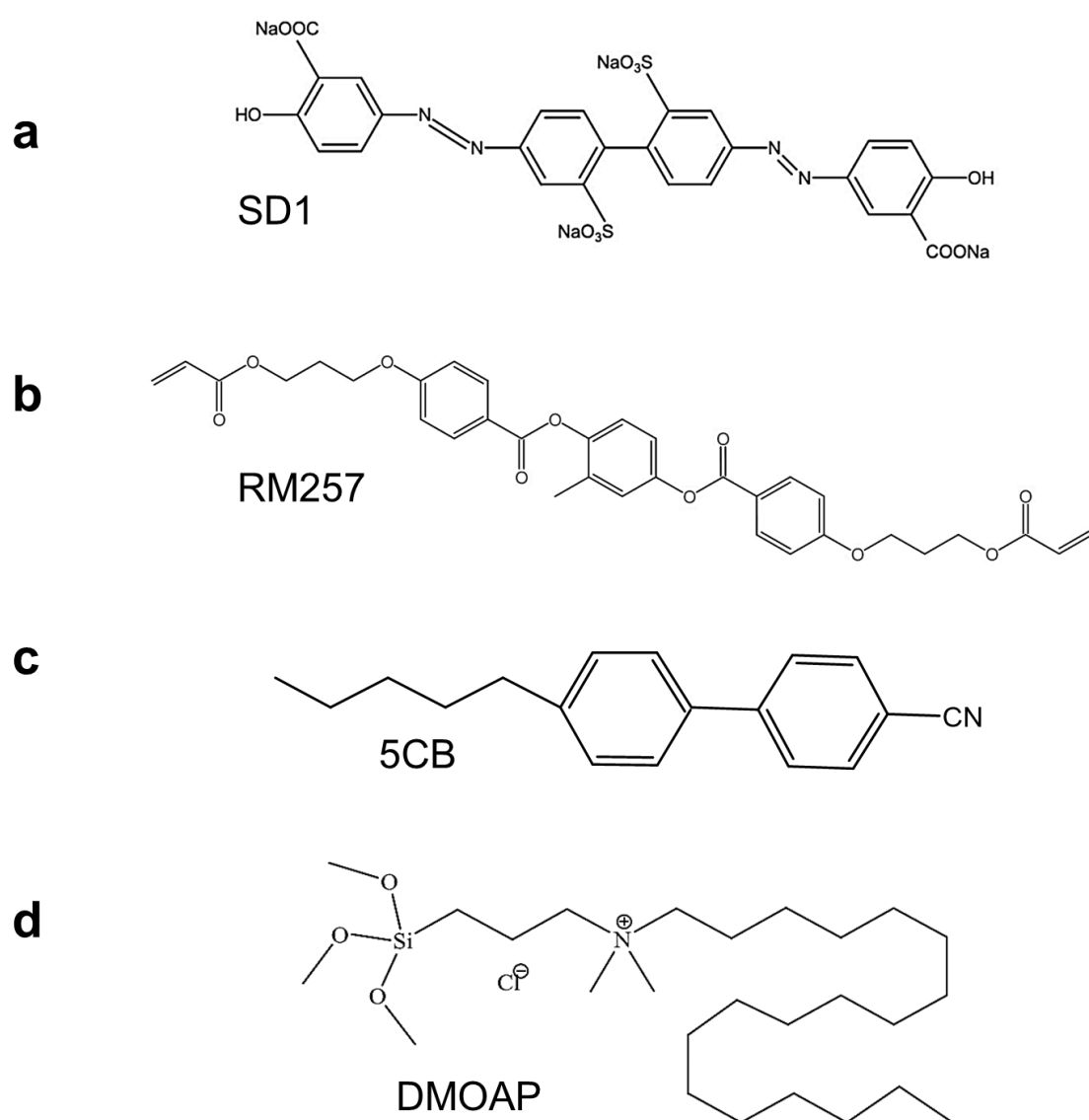
[†] J. Z., F. V. and Z. A. contributed equally to this work.

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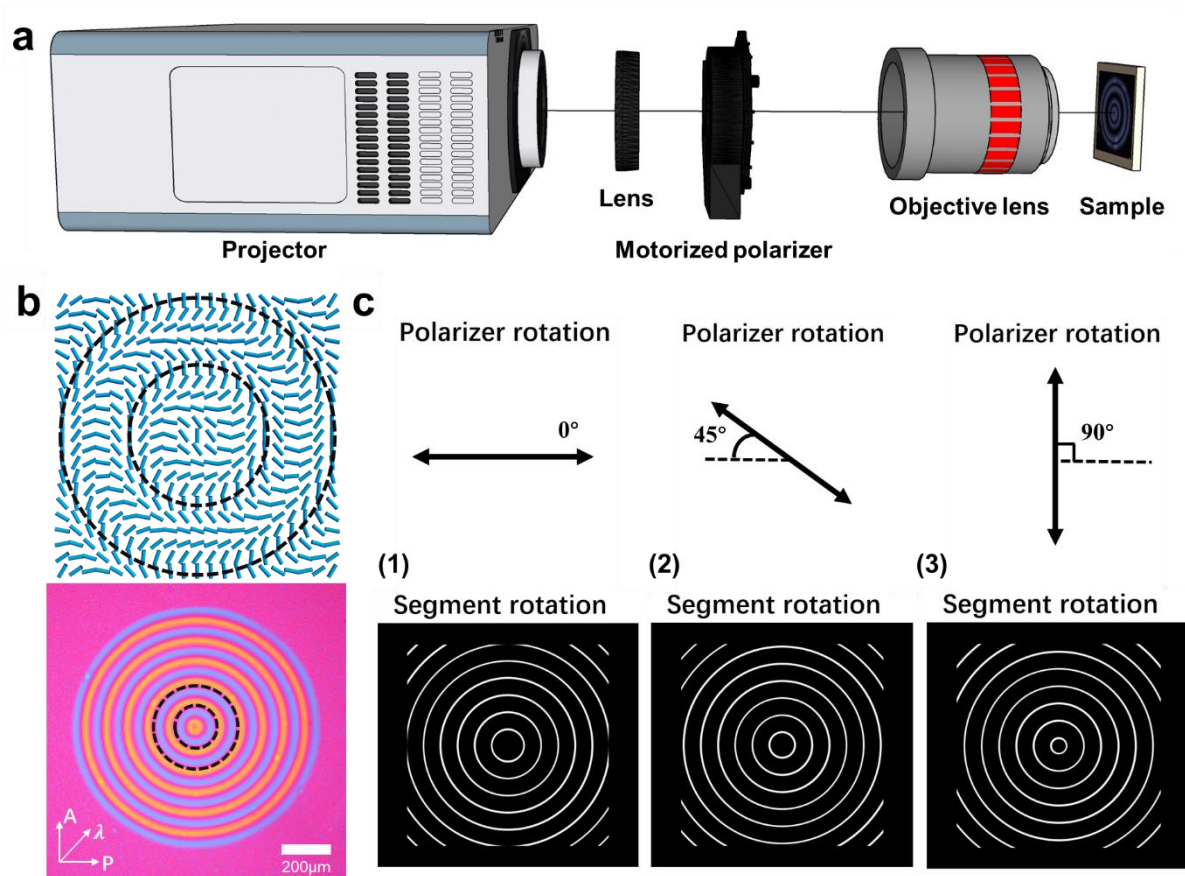
Supplementary Figures 1 to 14

Other Supplementary Materials for this manuscript include the following:

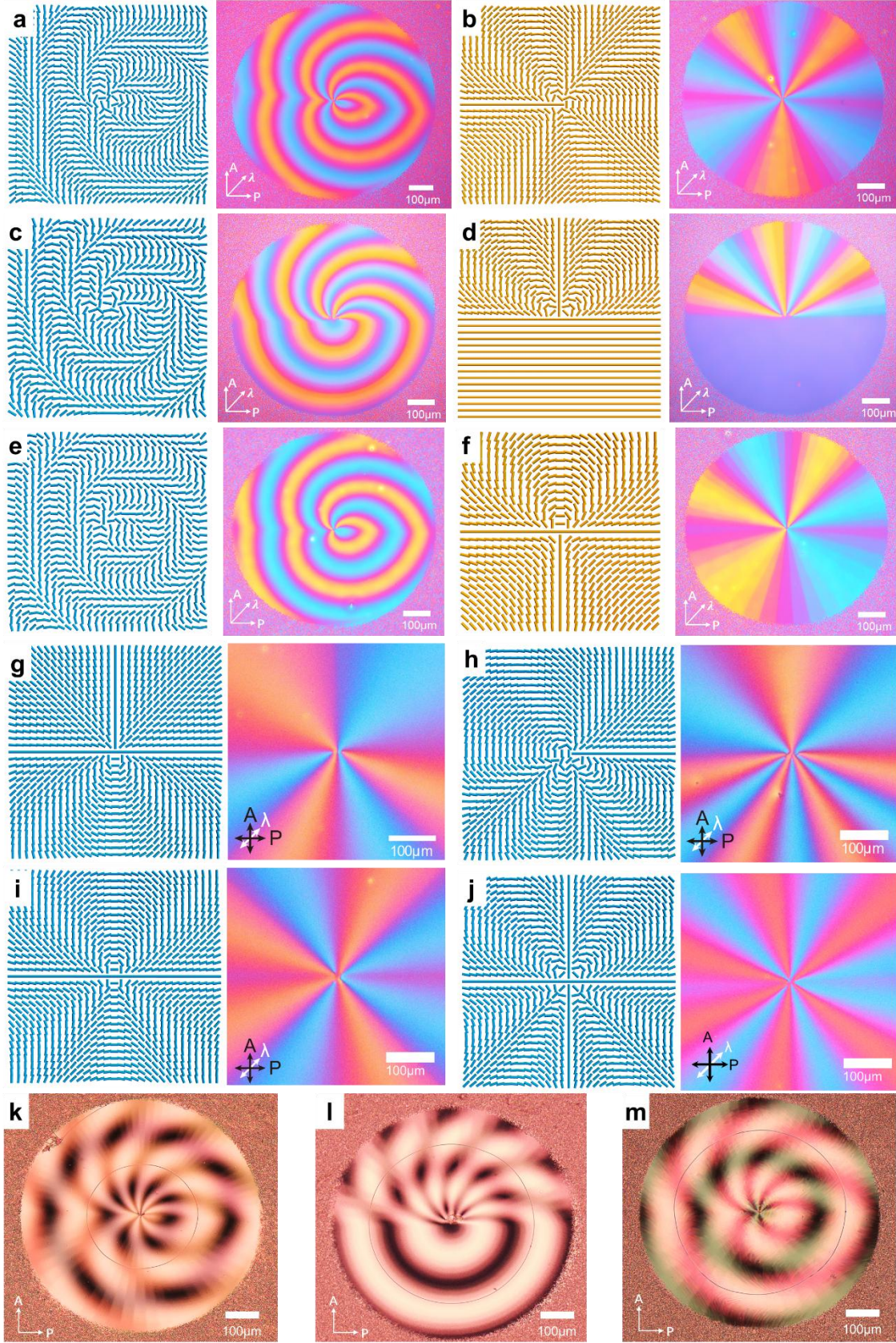
Supplementary Movies 1 to 14



Supplementary Fig. 1. Molecular structure of materials. (a) Azobenzene SD1 molecule; (b) Liquid crystal monomer RM257. (c) Nematic LC 5CB; (d) DMOAP;

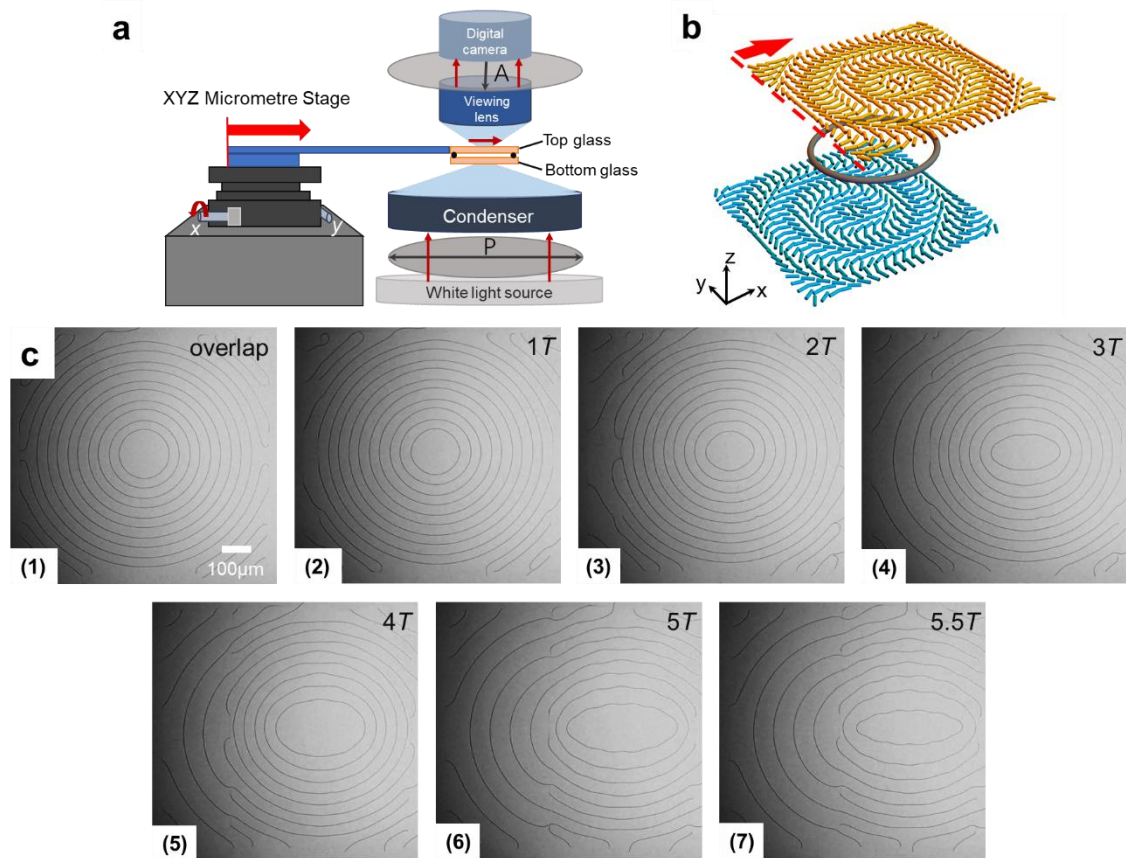


Supplementary Fig. 2. Maskless photopatterning setup. (a) Schematic of photopatterning setup; (b) The director field of the designed circular pattern and the corresponding polarizing optical microscope (POM) image with inserted red plate. (c) The relationship between the segment motion of the pattern and the rotation angle of the polarizer as it varies over time.

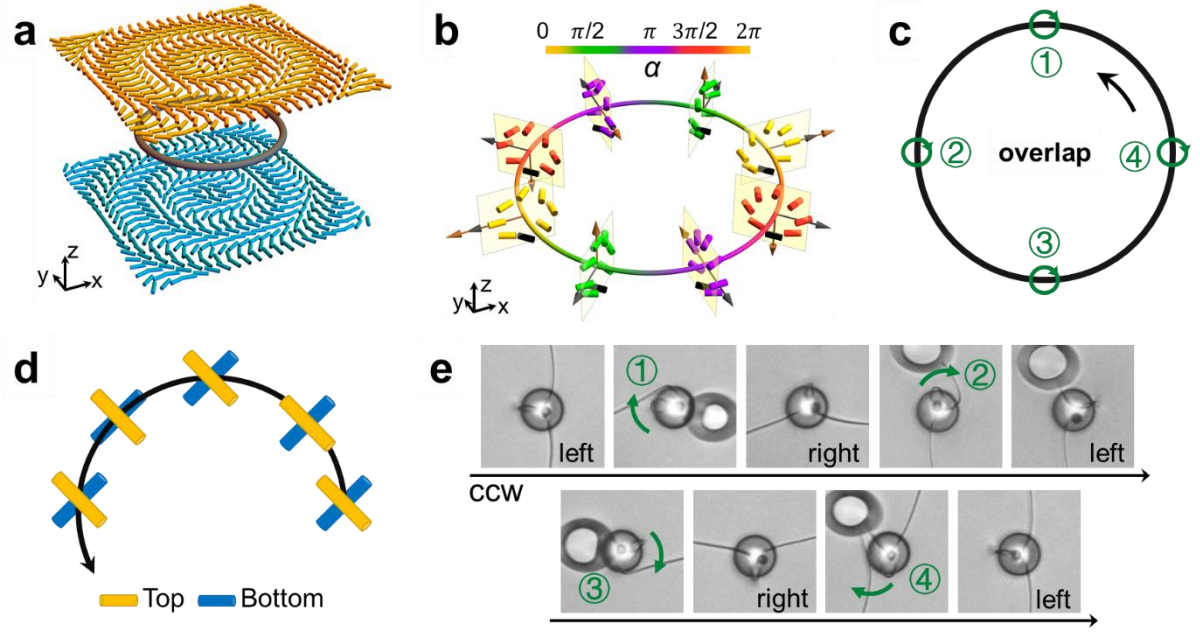


Supplementary Fig. 3. Asymmetric topological patterns used in this work. Director field of the designed pattern and the corresponding POM image with inserted red plate. (a) $\hat{n} = (n_x, n_y, n_z) =$

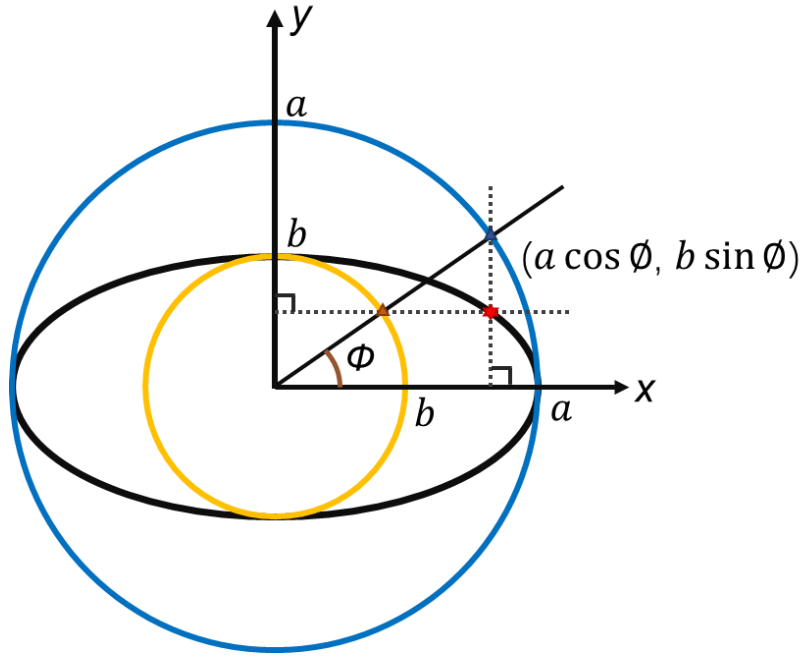
$(\cos\psi, \sin\psi, 0)$, $\psi = 2.5 \times \arctan \frac{y}{x} + \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y > 0$), and $\psi = -1.5 \times \arctan \frac{y}{x} + \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y < 0$). **(b)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = \frac{\pi}{2} + 2.5 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = \frac{\pi}{2} - 1.5 \times \arctan \frac{y}{x}$ ($y < 0$). **(c)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 3 \times \arctan \frac{y}{x} + \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y > 0$), and $\psi = \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y < 0$). **(d)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 3 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = 0$ ($y < 0$). **(e)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 2 \times \arctan \frac{y}{x} + \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y > 0$), and $\psi = -\arctan \frac{y}{x} + \frac{\pi\sqrt{x^2+y^2}}{T}$ ($y < 0$). **(f)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 2 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = -\arctan \frac{y}{x}$ ($y < 0$). **(g)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = \arctan \frac{y}{x}$ ($y > 0$), and $\psi = 2 \times \arctan \frac{y}{x}$ ($y < 0$). **(h)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 1.5 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = 3.5 \times \arctan \frac{y}{x}$ ($y < 0$). **(i)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 2 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = -2 \times \arctan \frac{y}{x}$ ($y < 0$). **(j)** $\hat{\mathbf{n}} = (n_x, n_y, n_z) = (\cos\psi, \sin\psi, 0)$, $\psi = 3 \times \arctan \frac{y}{x}$ ($y > 0$), and $\psi = -3 \times \arctan \frac{y}{x}$ ($y < 0$). **(k-m)** Polarizing optical microscope (POM) images of the disclination loop in the assembled sample. **(k)** The disclination loop in the sample is assembled by the designed bottom pattern in **(a)** and top pattern in **(b)**. **(l)** The disclination loop in the sample is assembled by the designed bottom pattern in **(c)** and top pattern in **(d)**. **(m)** The disclination loop in the sample is assembled by the designed bottom pattern in **(e)** and top pattern in **(f)**.



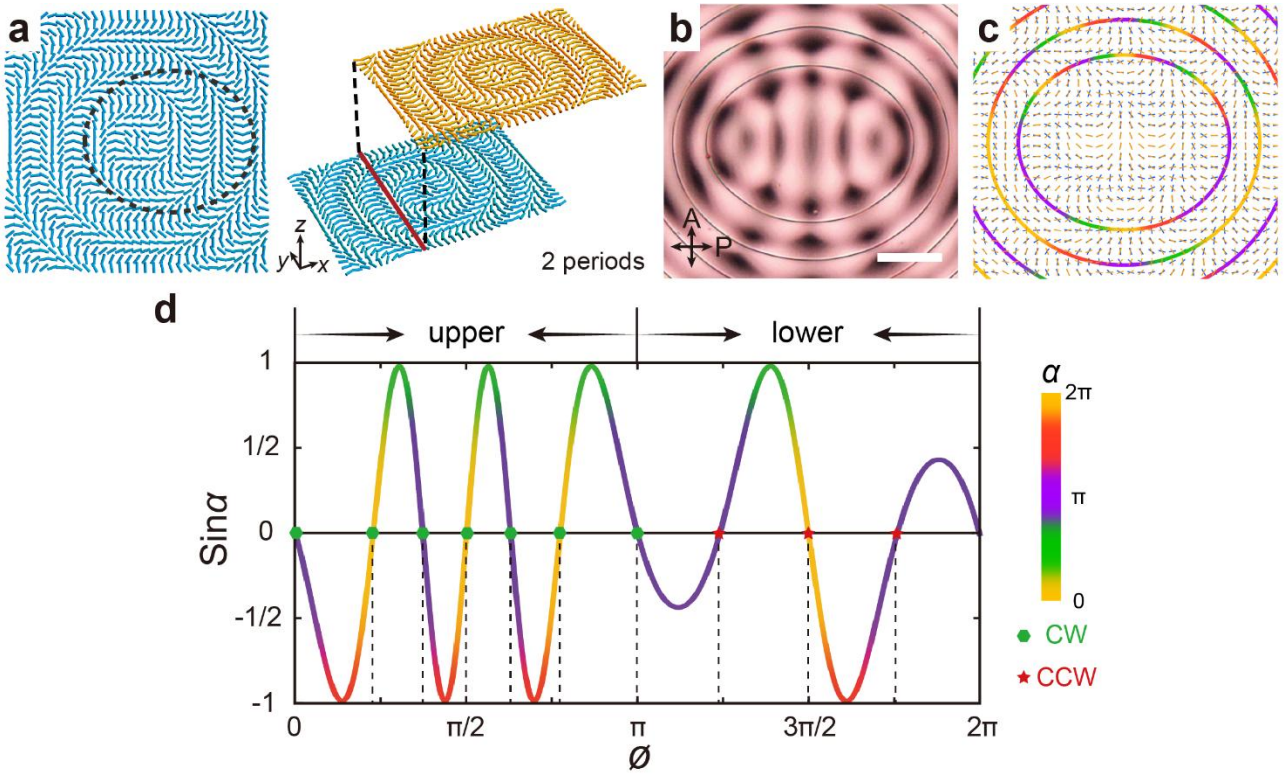
Supplementary Fig. 4. Geometric reconfiguration of the disclination loops during translation. (a) Schematic of the experimental setup. The top glass of the sample is pushed by a long glass slide mounted on the XYZ translation stage. Rotating the X-axis knob at a constant speed enables lateral translation of the pattern on the upper glass surface. (b) Schematic illustration of the top pattern shift in the sample. The pattern is translated along the $+x$ axis. (c) Image sequence of the geometric reconfiguration of the disclination loops during translation of the top pattern by 5.5 periods from the fully overlapped state. All translations are performed at a low speed to eliminate noticeable shear flow.



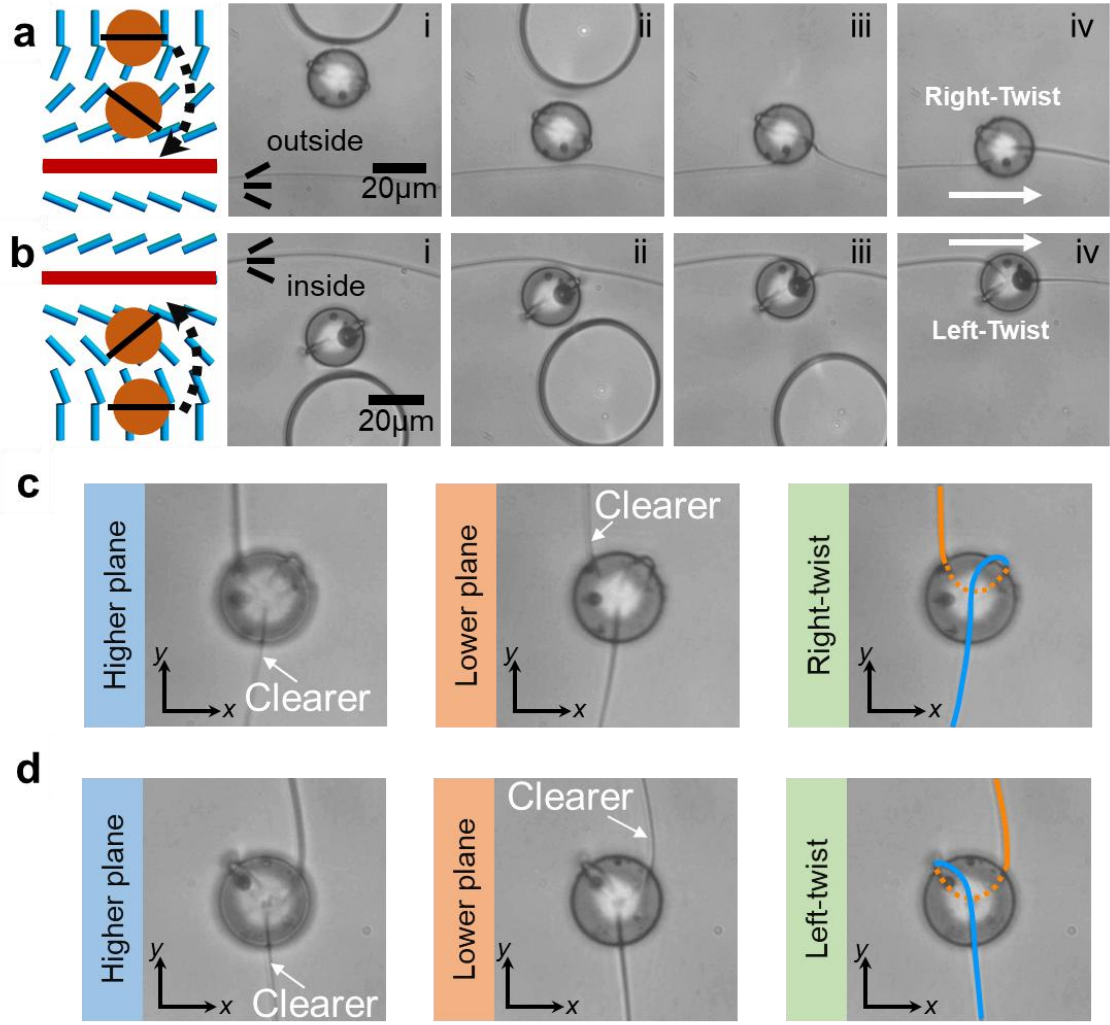
Supplementary Fig. 5. Symmetric rotation of entanglement in the disclination loop when the top pattern is overlapped. (a) Schematic of formation of disclination loop when the top and bottom patterns are totally overlapped. (b) The topological profile changes continuously along the loop and is central-symmetric. (c) Schematic of the symmetric rotation of the entanglement along the loop in CCW direction. (d) Schematic of the relative rotation of the top and bottom directors in the curved line loop. (e) Image sequence of rotation and chirality conversion of the colloidal entanglement. When the entanglement is moved along the loop in CCW direction, it rotates in CW direction 4 times and changes the chirality every rotation. The diameter of the particle is $d = 20 \mu\text{m}$.



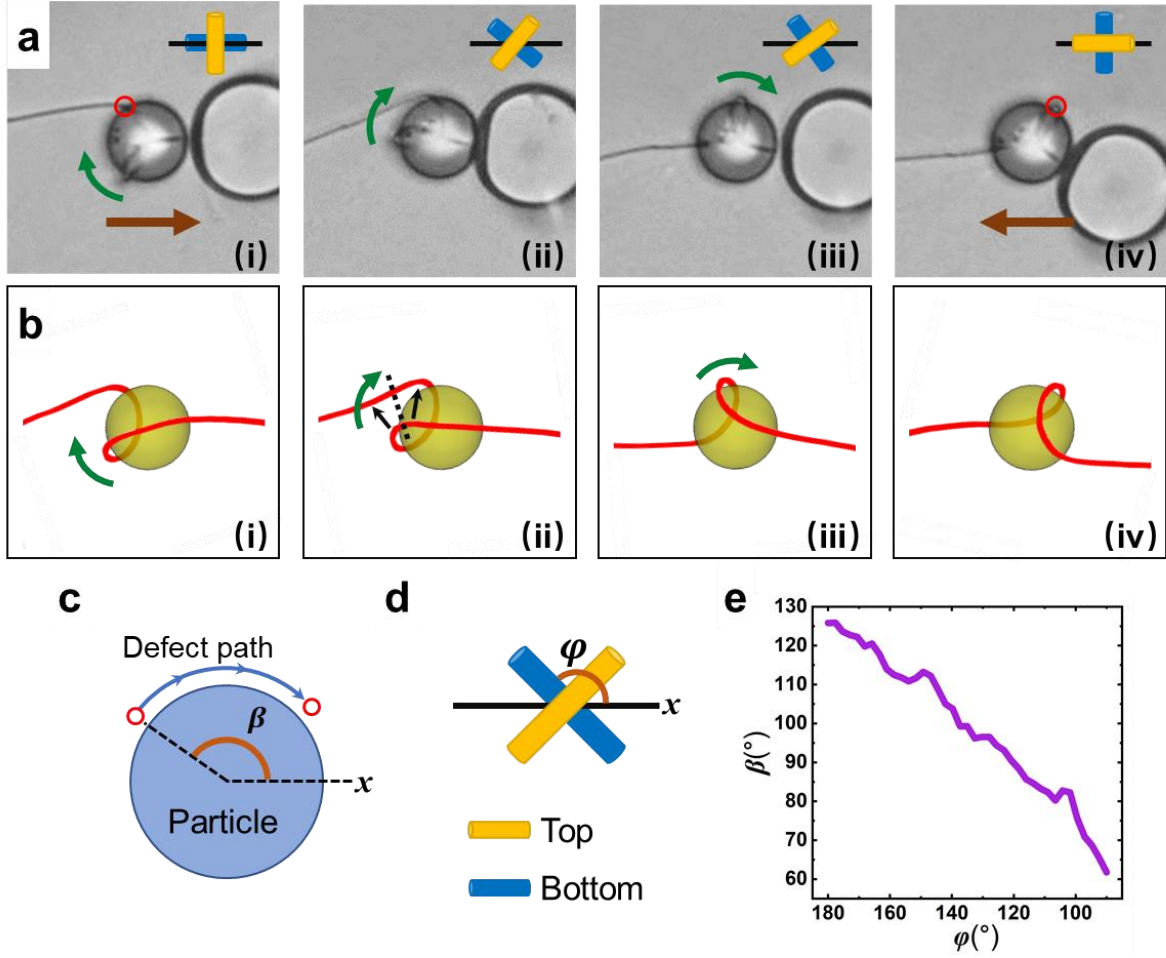
Supplementary Fig. 6. Schematic illustration of the angle ϕ . The elliptical disclination loops can be described by its major and minor axes as well as the parametric angle ϕ . The relation between the angle ϕ and the corresponding point in the loop is shown. The yellow circle has a radius equal to the minor axis of the ellipse, and the blue circle has a radius equal to the major axis of the ellipse. The corresponding point in the ellipse is $(a \cos \phi, b \sin \phi)$.



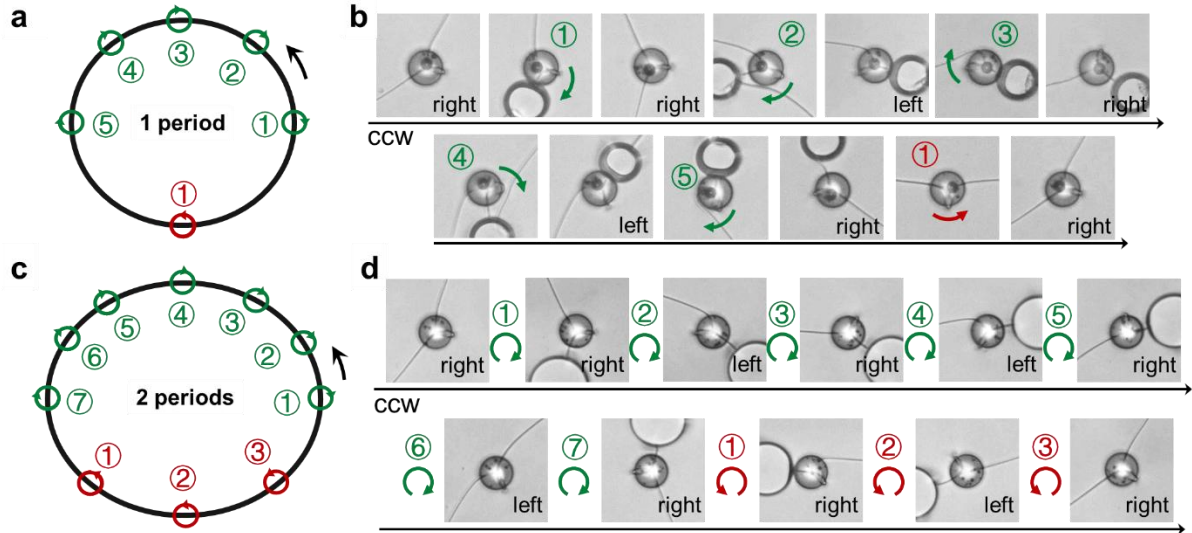
Supplementary Fig. 7. Topological profiles of elliptical disclination loops with the top pattern shifted by two periods. (a) Schematic of disclination loop formation with the top pattern shifted by two periods. (b) POM image of the corresponding disclination loop. (c) The change in angle α in the x-y plane after a two-period shift of the top pattern. (d) Correspondence between $\sin \alpha$ and the parameter angle $\emptyset$ of the disclination loop at two periods translation of the top surface. Seven points exhibit $\sin \alpha = 0$ on the upper half, and three points on the lower half. The scale bar in (b) is 100 μm .



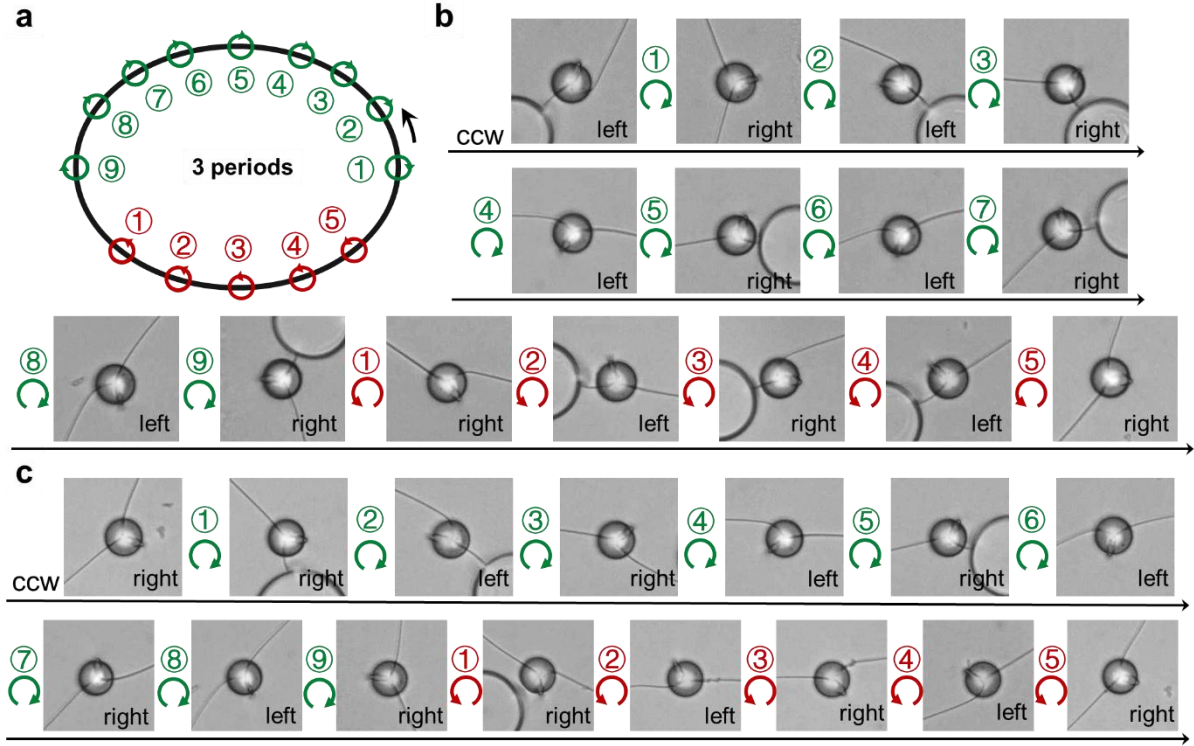
Supplementary Fig. 8. Formation of chiral entanglements and identification of their chirality. (a) Formation of right-twist colloidal entanglement from the outside interaction. (b) Formation of left-twist colloidal entanglement from the inside interaction. (c) Bright field images of the right-twist entanglement at different height. (d) Bright field images of the left-twist entanglement at different height.



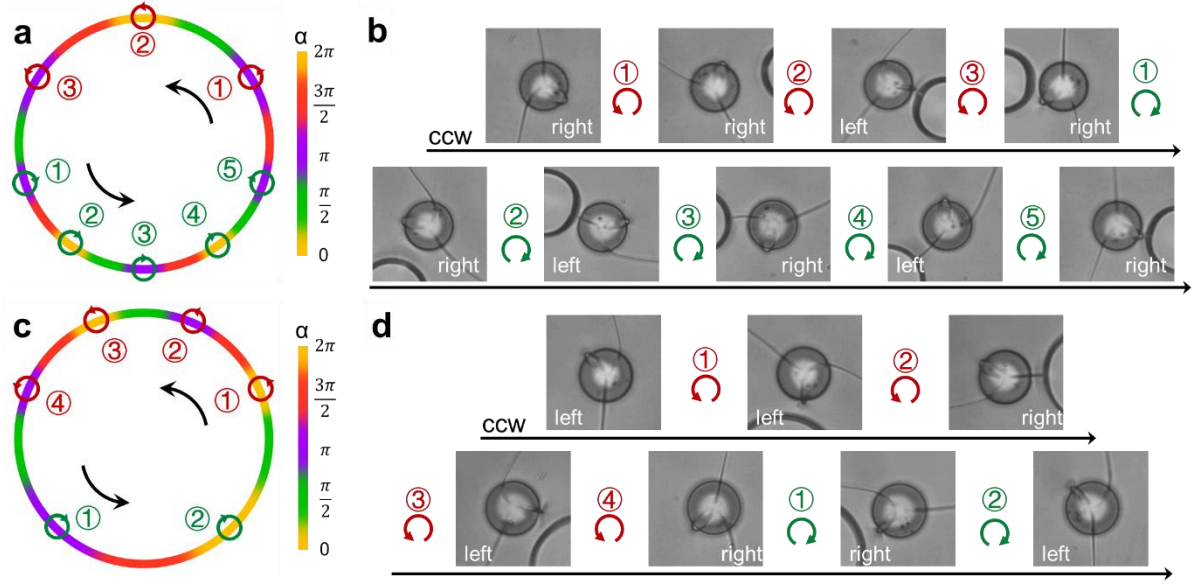
Supplementary Fig. 9. Dynamics of one colloidal entanglement rotation. (a) Image sequence of the entanglement rotation in experiments. The brown arrow points from the tangle to the colloid, and the corresponding top and bottom directors during rotation process are also shown in the upper right corner of each image. The black lines indicate the direction of the disclination line. (b) Corresponding image sequence of the entanglement rotation in simulation. The black arrows represent the relinking direction. (c) The entanglement rotates around the particle in CW direction, characterized by an angle β . (d) The definition of the angle φ . (e) The change of angle β with the angle φ .



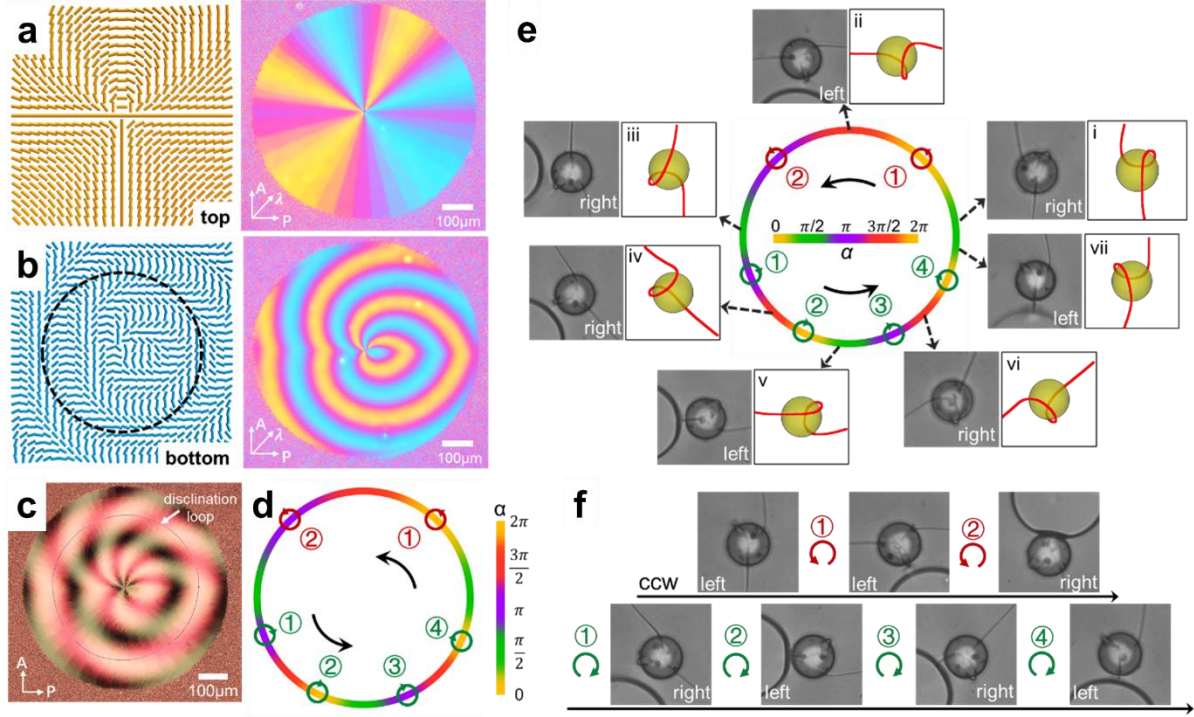
Supplementary Fig. 10. Another asymmetric chirality conversion of colloidal entanglement for top surface being shifted one and two periods. (a) Schematic of the asymmetric rotation of the entanglement along the loop in CCW direction for top surface being shifted one period. The entanglement will rotate in CW direction for 5 times at the upper half of the loop and rotate in CCW for only once at the lower half of the loop. (b) Image sequence depicting the rotation and chirality conversion of colloidal entanglement as it continues to move CCW for one cycle following the motion described in the main text. The entanglement starts with a chirality of right-twist and rotates in CW sense for 5 times from the position green ① to green ⑤. In the first rotation at the position green ①, it remains its chirality of right-twist. And it changes its chirality every time in the later 4 times rotation. However, it remains its chirality of right-twist when it rotates in CCW sense in the lower half of the loop. Thus, the entanglement changes its chirality 4 times in the upper half and 0 time in the lower half. And its final chirality of right-twist is the same with its initial state. (c) Schematic of the asymmetric rotation of the entanglement along the loop in CCW direction for top surface being shifted two periods. (d) Image sequence depicting the rotation and chirality conversion of colloidal entanglement as it continues to move CCW for one cycle following the motion described in the main text. The entanglement changes its chirality 6 times in the upper half and 2 times in the lower half. And its final chirality of right-twist is the same with its initial state. The diameter of the particle is $d = 20 \mu\text{m}$.



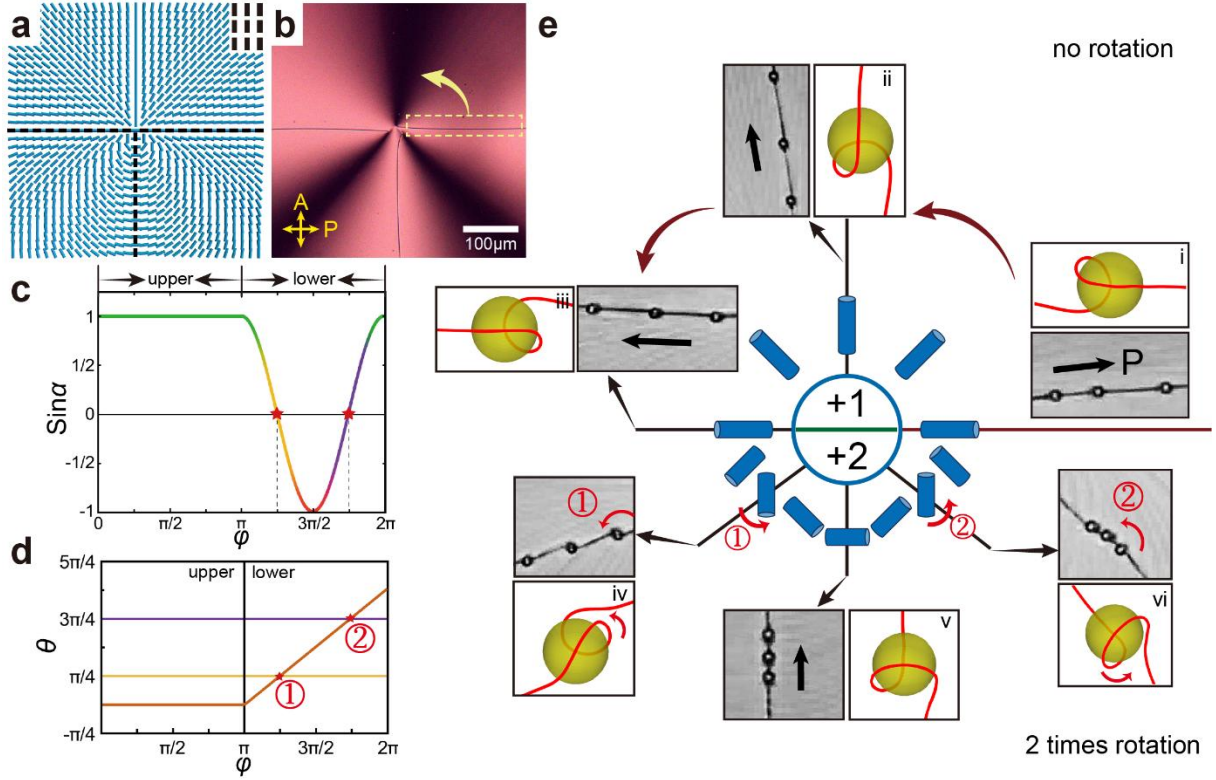
Supplementary Fig. 11. Two kinds of asymmetric chirality conversion of colloidal entanglement for top surface being shifted three periods. (a) Schematic of the asymmetric rotation of the entanglement along the loop in CCW direction for top surface being shifted three periods. The entanglement will rotate in CW direction for 9 times at the upper half of the loop and rotate in CCW for 5 time at the lower half of the loop. (b) Image sequence of rotation and chirality conversion of the colloidal entanglement. The entanglement starts with a chirality of left-twist and rotates in CW sense for 9 times from the position green ① to green ⑨. During this process, the entanglement changes its chirality every rotation. However, it remains its chirality of right-twist when it firstly changes the rotation direction to CCW sense at the position red ①. In the later 4 rotaion, it changes its chirality every time. The entanglement changes its chirality 9 times in the upper half and 4 times in the lower half. And its final chirality of right-twist is different with its initial state of left-twist. (c) Image sequence of rotation and chirality conversion of the colloidal entanglement continuing to move CCW for one circle. With the initial chirality of right-twist, the entanglement changes its chirality 8 times in the upper half and 4 times in the lower half. And its final chirality of right-twist is the same with its initial state. The diameter of the particle is $d = 20 \mu\text{m}$.



Supplementary Fig. 12. Another two programmable asymmetric chirality conversion of the colloidal entanglement. (a) 2D variation of the alpha angle and the schematic of the rotation. In this designed loop the entanglement will rotate in CCW direction for 3 times at the upper half of and rotate in CW for 5 times at the lower half of the loop. (b) Image sequence of rotation and chirality conversion of the colloidal entanglement. The entanglement starts with a chirality of right-twist, and it changes its chirality 2 times in the upper half and 4 times in the lower half. (c) 2D variation of the alpha angle and the schematic of the rotation. In this designed loop the entanglement will rotate in CCW direction for 4 times at the upper half of the loop and rotate in CW for 2 times at the lower half of the loop. (d) Image sequence of rotation and chirality conversion of the colloidal entanglement. The entanglement starts with a chirality of left-twist, and it changes its chirality 3 times in the upper half and 1 time in the lower half. The diameter of the particle is $d = 20 \mu\text{m}$.



Supplementary Fig. 13. Programmable asymmetric rotation and chirality conversion of entanglement. (a) Director field of the designed top pattern and its POM image with inserted red plate. (b) Director field of the designed bottom pattern and its POM image with inserted red plate. A disclination loop will form at the position of the dotted circle. (c) POM image of the sample featuring a disclination loop. (d) 2D variation of the alpha angle and the schematic of the rotation. (e) Experimental and simulation results for the rotation of colloidal entanglement in the loop. The entanglement starts with a chirality of right-twist. It rotates in CCW sense and changes its chirality at the position of red ① and ②. In the lower half of the loop, the entanglement remains its chirality of right-twist when it first changes the rotation direction to CW sense, and it changes the chirality every time in the later 3 CW sense rotations. (f) Image sequence depicting the rotation and chirality conversion of colloidal entanglement as it continues to move CCW for one cycle. The entanglement starts with a chirality of left-twist, and it changes its chirality once in the upper half and 3 times in the lower half.



Supplementary Fig. 14. Programmable non-equilibrium asymmetric dynamics of colloidal entanglement under a full cycle of disclination motion. (a) Schematic illustration of disclination lines formation. The bottom substrate features a designed pattern, and the top surface adopts a uniform direction along the y-axis. Disclination lines form at the position of the black dotted lines. (b) POM image of the disclination lines. (c) Variation of $\sin \alpha$ when the disclination line rotates to different azimuthal angle ϕ . (d) Variation of θ when the disclination line rotates to different azimuthal angle ϕ . (e) Experimental and numerical results on non-equilibrium asymmetric dynamics of colloidal entanglement during a full cycle of disclination motion. The entanglement starts with right-twist chirality (i). In the upper half of the pattern, it does not rotate. In the lower half, it rotates CCW two times from region red ① to red ②. The entanglement reverses its chirality after each rotation.