Description of Additional Supplementary Files

File name: Movie S1

Description: The thermoresponsive deformations of LCE microspheres and anisotropically-assembled structure. This video shows the top-view and side-view of thermally actuated LCE microspheres with a transition from spherical shape to pancake shape. Moreover, high-order architectures with more complex deformations can be achieved by anisotropic assembly of isotropic LCE microspheres.

File name: Movie S2

Description: The thermoresponsive deformations of LCE micro-ellipsoids. This video presents the thermoresponsive deformations of two LCE micro-ellipsoids with the alignment angle of θ = 0° and 90°, respectively, which show expansion and contraction along different directions.

File name: Movie S3

Description: The thermoresponsive deformations of LCE micro-rods. This video presents the thermoresponsive deformations of two LCE micro-rods with an alignment angle of θ = 45° for twisting actuation and a gradient in the mesogenic alignment for bending actuation, respectively.

File name: Movie S4

Description: The thermoresponsive deformations of helical LCE microfibers. The video presents the unwinding deformations of two helical LCE microfibers with the alignment angle of θ = 90° and 0°, respectively.

File name: Movie S5

Description: The thermoresponsive actuation of square-frame LCE architectures. This video includes three shape transformations with negative Poisson's ratios using square-frame LCE architectures with engineered molecular anisotropies. The first architecture is assembled using LCE micro-rods with $\theta = 0°$, displaying isotropic contraction; the second one is assembled using LCE micro-rods with $\theta = 90°$ and 90°, displaying anisotropic contraction; the third one is assembled using LCE micro-rods with a gradient in the mesogenic alignment, displaying frame bending.

File name: Movie S6

Description: The thermoresponsive actuation of a square-wave LCE architecture. The square-wave architecture is constructed by alternately assembling two types of LCE building blocks with well-aligned and poorly-aligned mesogens. The deformation occurs only at the amplitude of the square-wave architecture which constitutes LCE micro-rods with well-aligned mesogens.

File name: Movie S7

Description: The thermoresponsive actuation of a hexagonal LCE wheel. The hexagonal wheel consists of 6 well-aligned (as edge) and 6 poorly-aligned (as spoke) LCE micro-rods, which displays a shape change from 2D to 3D configuration.

File name: Movie S8

Description: Self-regulated locomotion of the tetrahedral LCE microactuator. Actuated by a rotating magnetic field at a speed of 18 rpm (rounds per minute), the tetrahedral microactuator demonstrates self-regulated locomotion by spontaneously switching between rolling and flipping modes.

File name: Movie S9

Description: Magnetically-guided motion of tetrahedral LCE microactuators along predetermined trajectories. The tetrahedral microactuator moves along a square-shaped path and an "S"-shaped path guided by a magnetic field with self-regulated locomotion.

File name: Movie S10

Description: The tetrahedral microactuator climbing over obstacles. The tetrahedral microactuator successively climbs over three cylindrical obstacles with a height of 450 μm.

File name: Movie S11

Description: Static magnetic-guided translational motion of the tetrahedral microactuator. The microactuator undergoes translational motion along the direction of a static magnetic field in the horizontal plane. By sequentially altering the direction of the magnetic field, the microactuator follows trajectories forming "L", "□", and "C" shapes, respectively.

File name: Movie S12

Description: Magnetic leaping of the microactuator to pass an obstacle in liquid. The microactuator, immersed in 10 cSt viscosity silicone oil, experiences upward motion under the actuation of a static magnetic field positioned above it, successfully traversing an obstacle as the position of the magnetic field changes. Upon removal of the magnetic field, the microactuator descends back to the bottom surface of the container.

File name: Movie S13

Description: Manipulation of a solid cargo by the tetrahedral microactuator. The pre-folded microactuator traverses a serpentine microchannel (650 μm in width), then recovers its original structure (with a characteristic length of ~850 μm) when heated to 80 °C. Afterward, the microactuator catches a solid cargo (dyed red), transports it to a targeted location, and releases it.

File name: Movie S14

Description: Manipulation of liquid droplets by the tetrahedral microactuator. The tetrahedral microactuator captures an $AgNO_3$ (0.2 M, red) and a KCl (0.4 M, green) microdroplet successively. After coalescence, the two reagents are mixed in the heated droplet microreactor for chemical reaction. By rotating the microactuator magnetically at a speed of 300 rpm, the product gradually accumulates in the bottom of the microreactor. After that, the microactuator releases the droplet microreactor, in which an aggregation of silver nanoparticles (black) is obtained.