Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information

Design principles for non-reciprocal photomechanical actuation

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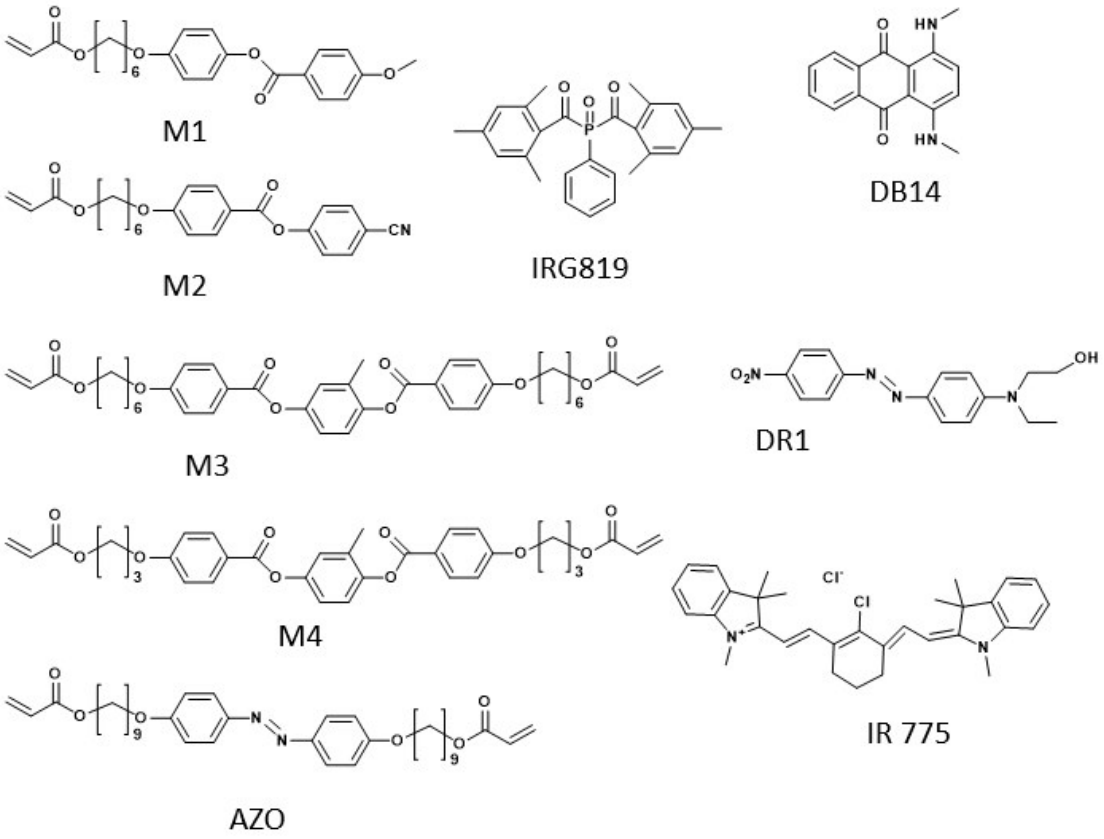

Scheme S1. Chemical structures of the molecules used.

Table S1. Names of the molecules:

M1	4-Methoxybenzoic acid 4-(6-acryloyloxyhexyloxy)phenyl ester
M2	4[4[6-Acryloxyhex-1-yl)oxyphenyl]carboxybenzonitrile
M3	1,4-Bis-[4-(6-acryloyloxyhexyloxy)benzoyloxy]-2-methylbenzene
M4	1,4-Bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene
Azo	4,4'-Bis[9-(acryloyloxy)nonyloxy]azobenzene
IRG819	Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide
DR1	*N*-Ethyl-*N*-(2-hydroxyethyl)-4-(4-nitrophenylazo)aniline
DB14	1,4-Bis(methylamino)anthraquinone
IR775	2-[2-[2-Chloro-3-[2-(1,3-dihydro-1,3,3-trimethyl-2H-indol-2-ylidene)-ethylidene]-1-cyclohexen-1-yl]-ethenyl]-1,3,3-trimethyl-3H-indolium chloride

Table S2. Chemical composition (in mol %) of the different actuators used.

	LCN-Azo	LCN-DB14	LCN-DR1	Self-oscillating
note	Photochemical	Photothermal	Photothermal	Photothermal
M1	52.6	52.6	52.7	78
M2	18.4	17.5	17.7	-
M3	21.5	27.5	27.6	-
M4		-	-	20
IRG 819	1.5	1.3	1.3	1
Azo	6	-	-	-
DB14	-	1.1	-	-
DR1	-	-	0.7	1

Table S3. The light sources used for actuation.

Control strategy	385 nm ($mW\ cm^{-2}$)	490 nm ($mW\ cm^{-2}$)	660 nm ($mW\ cm^{-2}$)	532 nm laser (mW)
3 wavelengths	8	30	100	-
2 wavelengths	8 (constantly)*	80	100	-
1 wavelength	8 (constantly)*	80	-	-
0 wavelength	-	-	-	100 (constantly)*

**Constantly* means a continuous light beam without spatial or temporal modulation.

Table S4. The light-control sequences for the different actuation schemes.

Strategy	3-wavelength modulation	2-wavelength modulation	1-wavelength modulation	0-wavelength modulation
Composition	LCN-DB14 LCN-Azo	LCN-DB14 LCN-Azo	LCN-DR1 LCN-Azo	LCN-DR1
Schematic				
Light control sequence	At 0 s, UV on	UV always on	UV always on	532 nm Laser on
	At 20 s, UV off, Red on	At 20 s, Red on	At 20 s, Blue on	Constantly
	At 25 s, Blue on	At 25 s, Blue on	At 30 s, Blue off	
	At 35 s, Red off, Blue off	At 35 s, Red off, Blue off	Repeat	
	Repeat	Repeat		

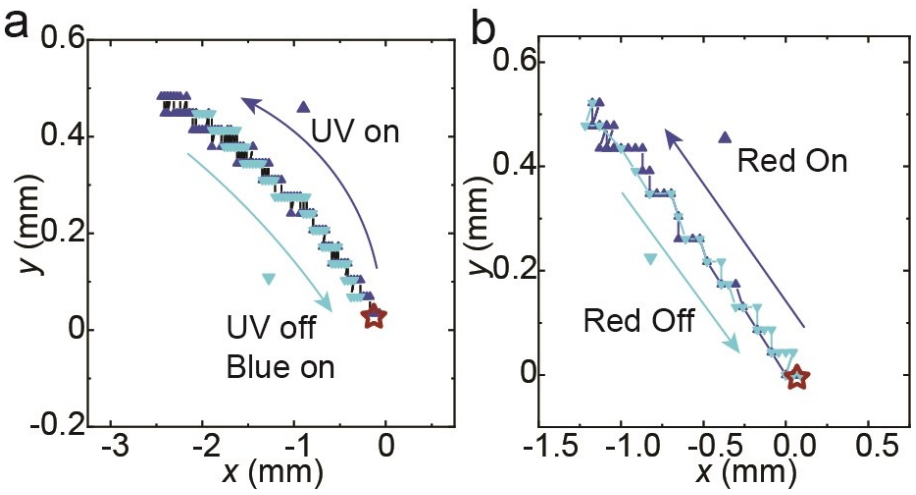

Fig. S1. Reciprocal motion by using only the photochemical (left) or photothermal (right) actuation. The hybrid, two-segment LCN strip is composed of both the photochemical and photothermal segments. (a) X-Y trajectory of the photochemical actuation, by illumination with UV (bending) and blue (unbending) light. UV: 385 nm, 20s, 8 mW cm^{-2}; Blue: 490 nm, 10s, 80 mW cm^{-2}. (b) X-Y trajectory of the photothermal actuation, by switching on/off the red-light excitation (660nm, 5s, 100 mW cm^{-2}).

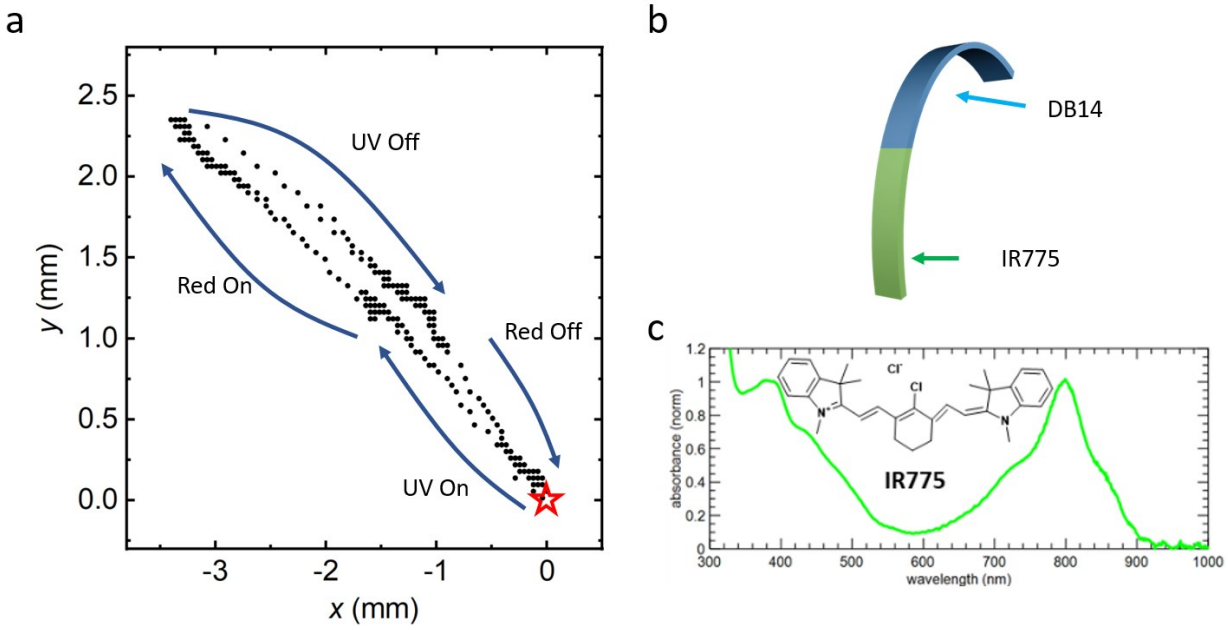

Fig. S2. Non-reciprocal motion of an LCN strip composed of two photothermal segments. (a) X-Y trajectory upon the two-light control sequence: (1) UV on (0 s); (2) Red on (5 s); (3) UV off (10 s); (4) Red off (15 s). (b) Schematic drawing of the two-segment construction of the actuator. c) Absorption spectrum of the UV absorbing dye used in the photothermal actuation of the bottom part. Inset: the chemical structure of the dye. IR775 concentration: 0.5 mol %. Light sources: 130 mw cm^{-2} (405 nm) and 50 mw cm^{-2} (660 nm).

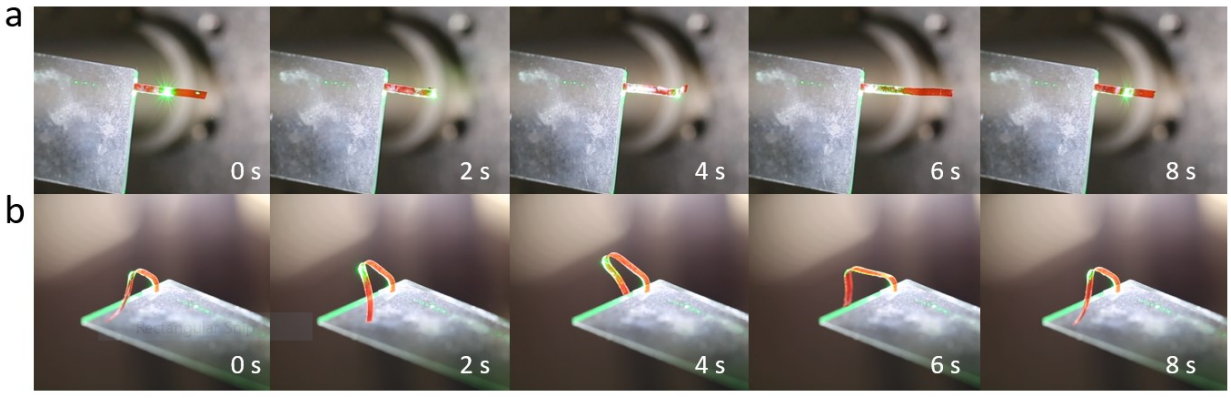

Fig. S3. Top view (a) and bottom view (b) photos during one non-reciprocal self-oscillation cycle. The spot position indicates that light excitation occurs only on the upper surface of the strip.

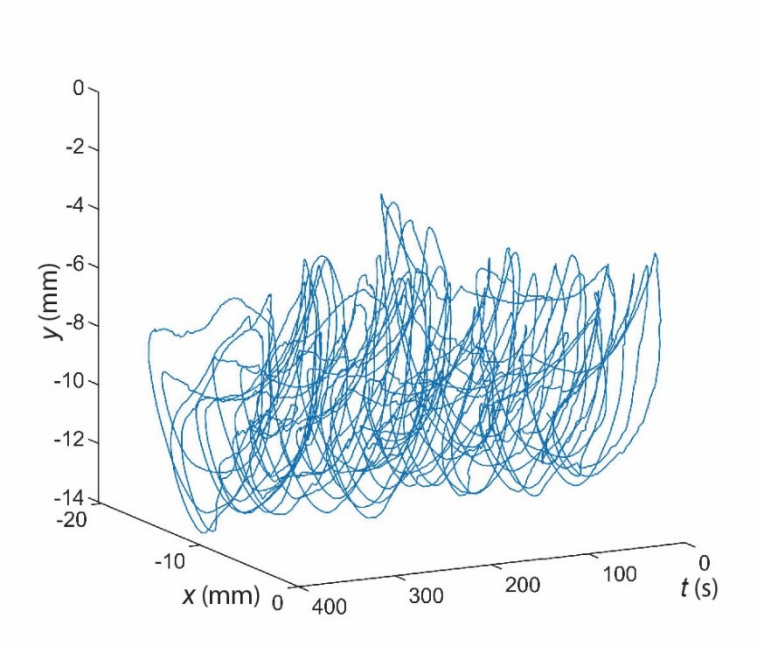

Fig. S4. Evolution of non-reciprocal trajectory on X-Y plane during 400 s oscillation.

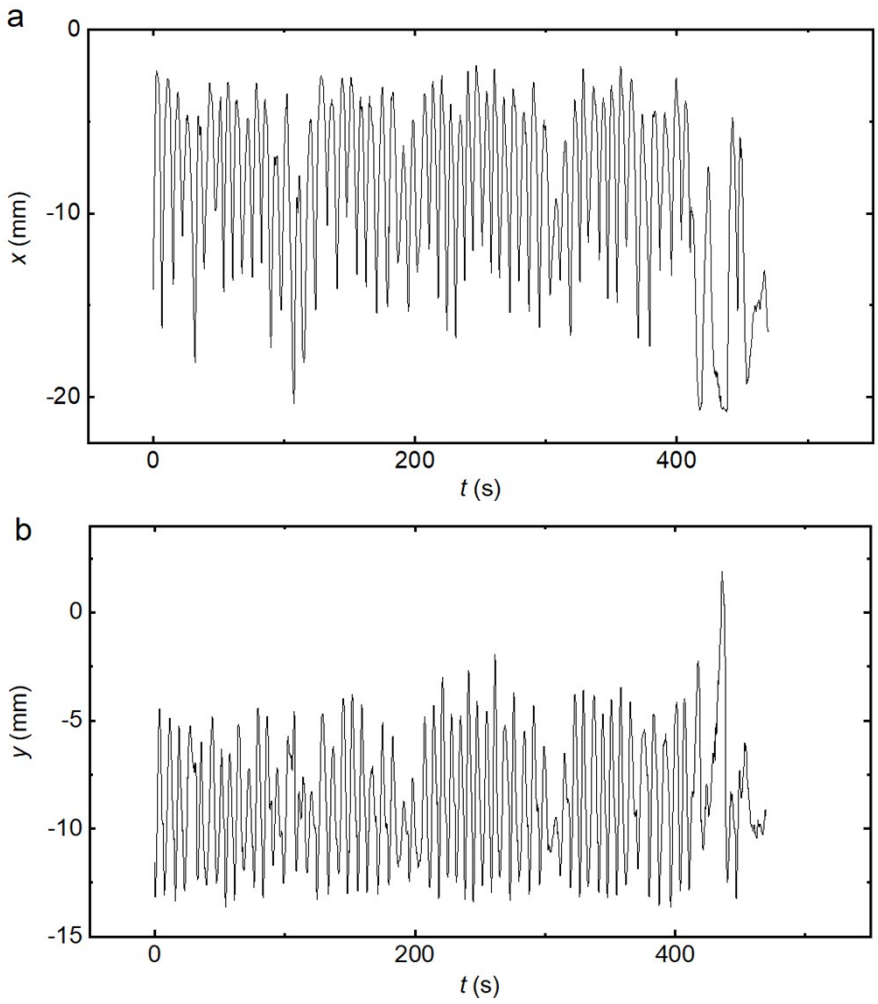

Fig. S5. Oscillation dynamics of the non-reciprocal self-oscillator. Data for tip displacement in X-axis (a) and Y-axis (b) during the oscillation.

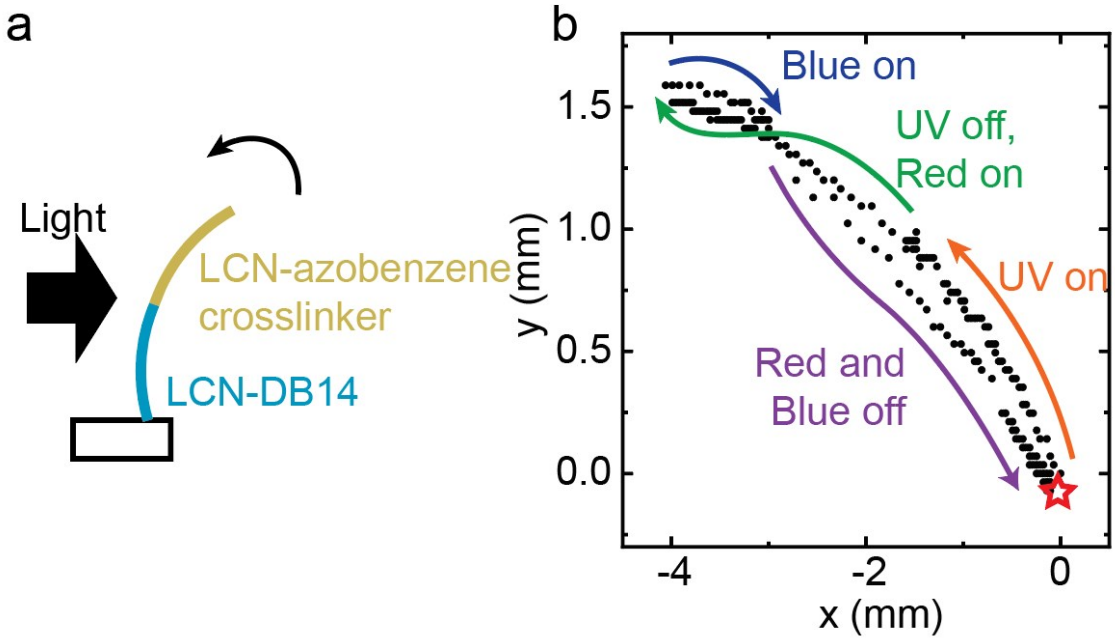

Fig. S6. Non-reciprocal motion of an LCN strip fixed such that the photothermal segment is at the bottom. (a) Schematic drawing of the two-segment construction of the actuator. (b) X-Y trajectory upon control sequence utilizing three light beams: (1) UV on (0 s); (2) UV off, Red on (20 s); (3) Blue on (25 s); (4) Red and Blue off (35 s). Light sources: 8 mW cm^{-2} (UV, 385 nm), 30 mW cm^{-2} (Blue, 490 nm) and 100 mW cm^{-2} (Red, 660 nm). Actuator dimensions: 15×1×0.03 mm^3.

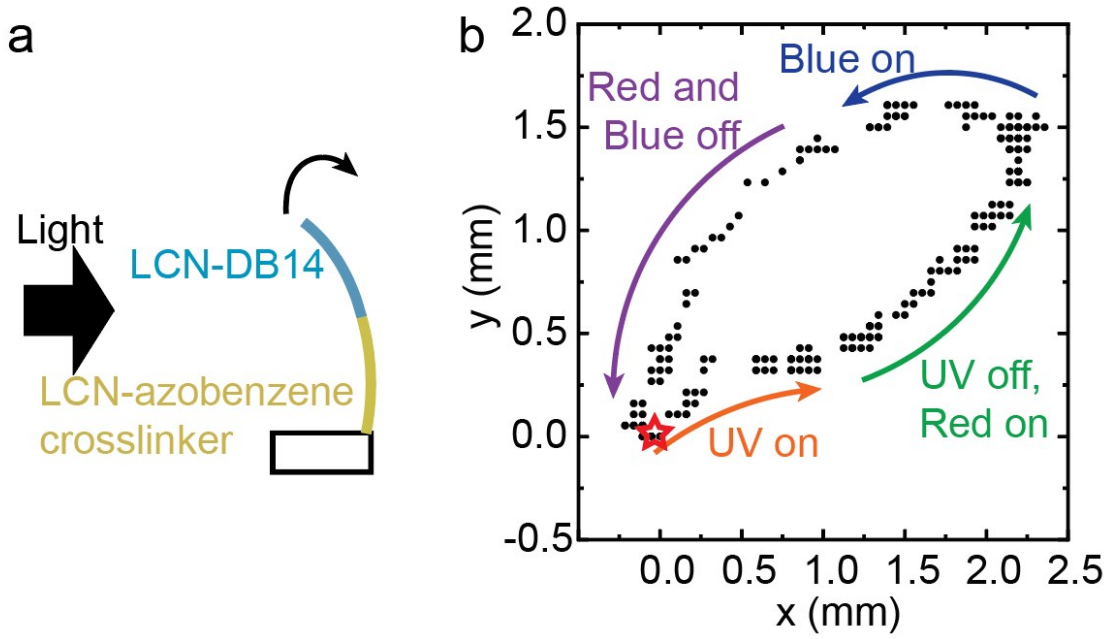

Fig. S7. Non-reciprocal motion of an LCN strip by reversing the bending direction. (a) Schematic drawing of the two-segment construction of the actuator in respect to the light incident and strip bending directions. (b) X-Y trajectory upon control sequence utilizing three light beams: (1) UV on (0 s); (2) UV off, Red on (20 s); (3) Blue on (25 s); (4) Red and Blue off (35 s). Light sources: 8 mW cm^{-2} (UV, 385 nm), 30 mW cm^{-2} (Blue, 490 nm) and 100 mW cm^{-2} (Red, 660 nm). Actuator dimensions: 15×1×0.03 mm^3.

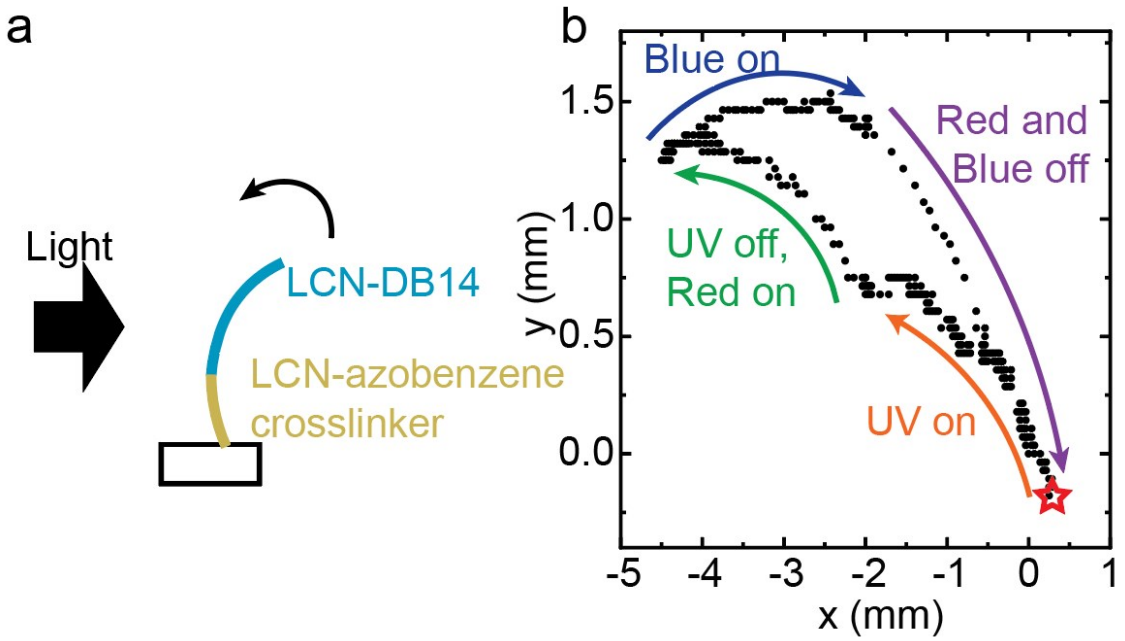

Fig. S8. Non-reciprocal motion of an LCN strip with unequal segment lengths. (a) Schematic drawing of the two-segment construction of the actuator. The length of LCN-DB14 is two times of LCN-Azo (azobenzene crosslinker). (b) X-Y trajectory upon control sequence utilizing three light beams: (1) UV on (0 s); (2) UV off, Red on (20 s); (3) Blue on (25 s); (4) Red and Blue off (35 s). Light sources: 8 mW cm^{-2} (UV, 385 nm), 30 mW cm^{-2} (Blue, 490 nm) and 100 mW cm^{-2} (Red, 660 nm). Actuator dimension: 15×1×0.03 mm^3.