**Description of Additional Supplementary Files**

File Name: Supplementary Movie 1

Description: Showing deformation reversal of aligned LCN1(20,120) actuator during one cooling process.

File Name: Supplementary Movie 2

Description: Deformation reversal of aligned LCN2(540%,400%), LCN2(700%,400%) and LCN2(1000%,400%) actuators during cooling.

File Name: Supplementary Movie 3

Description: Twisting-untwisting bidirectional deformation of a -45o aligned LCN1(60,120) actuator during cooling.

File Name: Supplementary Movie 4

Description: Unwinding-winding bidirectional deformation of a +45o aligned LCN actuator during cooling.

File Name: Supplementary Movie 5

Description: Light-induced deformation reversal behavior of polydopamine(PDA)-coated LCN actuators: PDA-LCN1(20,120) and PDA-LCN2(1000%,400%).

File Name: Supplementary Movie 6.

Description: Light-driven micro-walker of PDA-LCN1(20,120).

File Name: Supplementary Movie 7

Description:Llight-driven deformation and locomotion of a wave-shaped walker of PDA-LCN actuator.

File Name: Supplementary Movie 8.

Description: Light-driven multi-mode locomotion of a single PDA-LCN actuator enabled by deformation reversal upon different light irradiation patterns.