Supporting Information

Near-Infrared Light-Driven Locomotion of a Liquid Crystal Polymer Trilayer Actuator

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1. Materials. All solvents and reagents were purchased from Sigma Aldrich and used without further purification. Graphene oxide (GO), used to prepare reduced graphene oxide (RGO), was kindly provided by Prof. Hesheng Xia of Sichuan University (produced by The Sixth Element Materials Technology Co., Ltd, China).

2. General Characterizations. The cross-sectional morphology of trilayer actuators was examined using a Hitachi S-4700 field-emission-gun scanning electron microscope (SEM) operating at 1.0 kV to10.0 kV. For SEM observation, a fine platinum coating layer (a few nm) was laminated on sample surface by using a K550 sputter coater for 1 or 2 min. The bending/unbending angles of the actuators were measured by applying NIR laser (980 nm, normal incidence, light source 10 cm away) to a strip-shaped sample (15 mm × 2 mm × 94 μm). The isostrain measurement for Actuator-3 (experimental setup shown in Figure S4) was performed at room temperature under NIR (785 nm) light irradiation on an Instron 5965 system with a 5 N load cell using the stress-relaxation mode. By applying NIR light (normal incidence, light source 10 cm away) to a stripe-shaped sample held under constant strain (within 0.2 %), the contractile force generated in the film can be sensed and measured. The photothermally induced heating was monitored by measuring the temperature of the actuator using a thermal imager (Tseto 875i) with a temperature resolution of 0.1 °C. While applying the laser on the graphene top layer, the temperature on the other side (either RGO/polymer/LCN trilayer or RGO/polymer bilayer) was recorded. The NIR light intensity was measured using a photometer (Oriel® Instruments).

3. Preparation of RGO Layer. RGO was prepared according to a literature method.[1] In a typical procedure, the GO was firstly dispersed in water to create a 0.1 wt% dispersion by ultrasonication for 2 h. Then the resulting homogeneous dispersion was reduced with a hydrazine solution in a 95 °C oil bath for 1 h. The weight ratio of hydrazine to GO was about 7:10. RGO layers with different thicknesses were prepared by vacuum filtration of different volumes of RGO dispersions through a membrane filter (0.45 μm in pore size), which was followed by drying at room temperature for 6 hours. Finally, the RGO sheet was peeled off from the membrane for further use. In this work, three RGO sheets of varying thickness were obtained by using 5, 6 and 7ml of RGO dispersions, respectively.

4. Preparation of LCN layer and RGO/Acrylic Ester/LCN Trilayer Actuator. Fig. S1 schematically shows the fabrication process of the trilayer actuator. The LCN film was firstly stretched at 55 ^{o}C (nematic phase) to 300% elongation. Then the external force was removed and the two sides of the film were each exposed to UV light (320-480 nm filter, 90 mW/cm^2) for 30 min, reaching essentially uniform crosslinking of the polymer. Trilayer actuator was then assembled by connecting the RGO layer to the stretched LCN through an acrylic ester tape (purchased from Suzhou Tuojia Technology Co., Ltd, China) that becomes the inactive middle polymer layer.

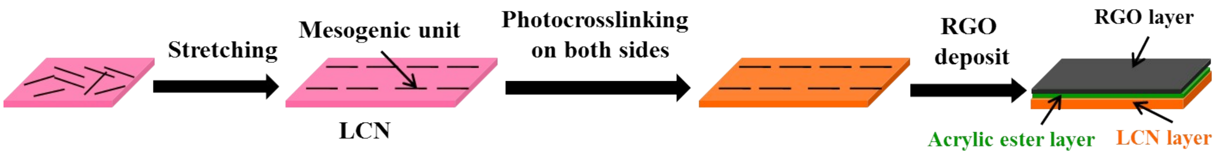

Fig. S1 Schematic illustration of the fabrication process of the RGO/Polymer/LCN trilayer actuator.

5. Additional Characterization Results

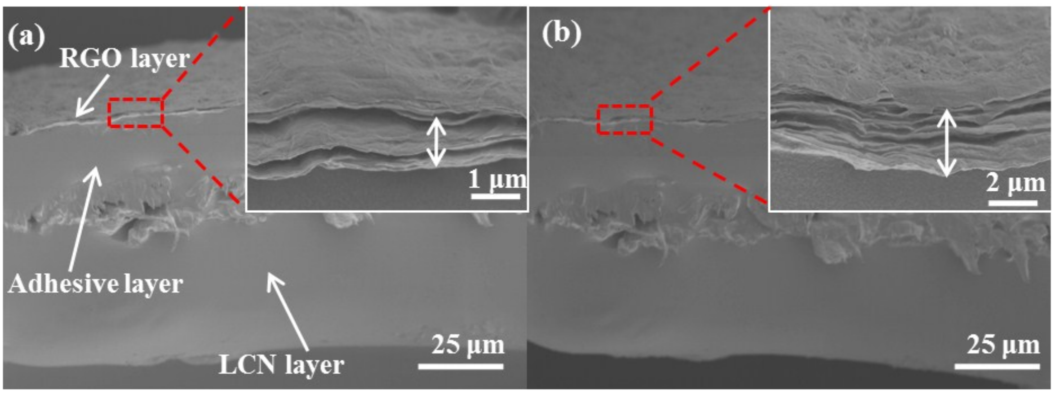

Fig. S2 Cross-sectional SEM image of the trilayer actuator-1 (a) and actuator-2, differing in the thickness of the RGO layer.

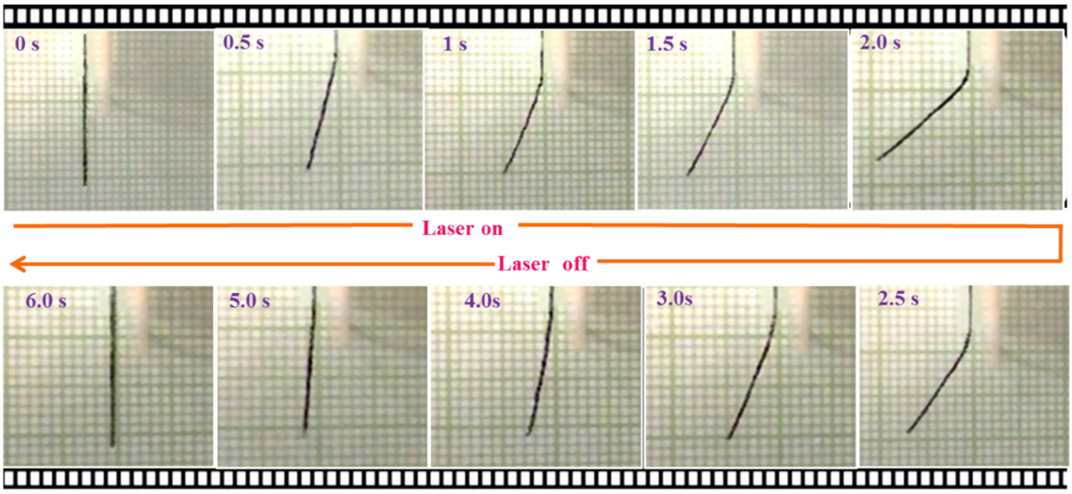

Fig. S3 Snapshots showing the reversible bending/unbending of the actuator-3 in response to NIR laser irradiation (1.87 W/cm^2) on/off, respectively.

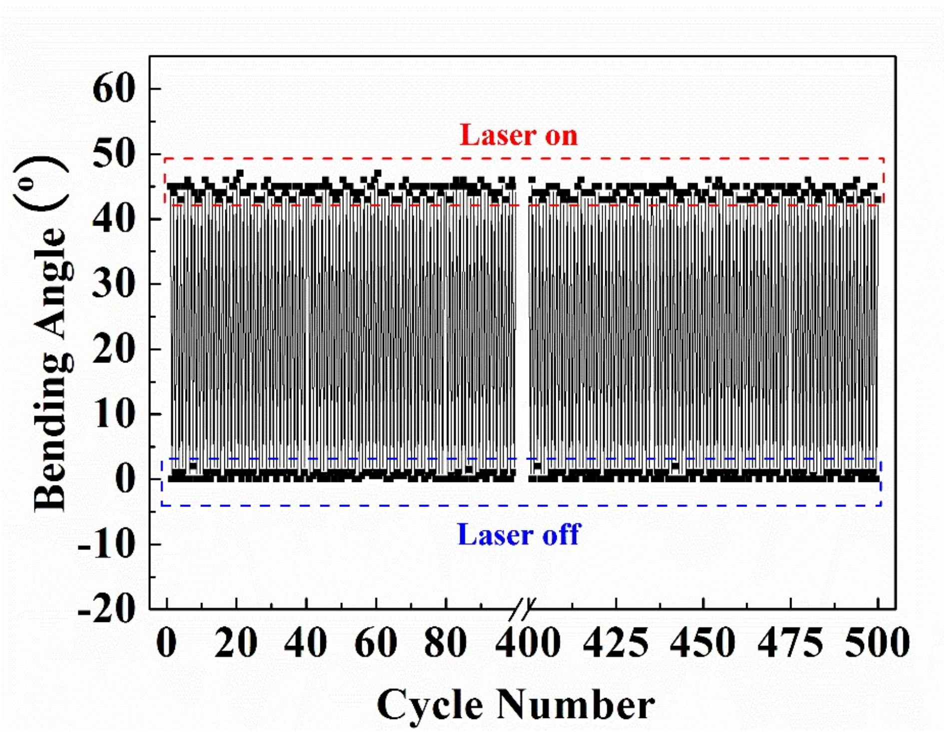

Fig. S4 Life cycle of actuator-3 subjected to 500 cycles of NIR light on for 2 seconds (1.87 W/cm^2) and off for 4 seconds.

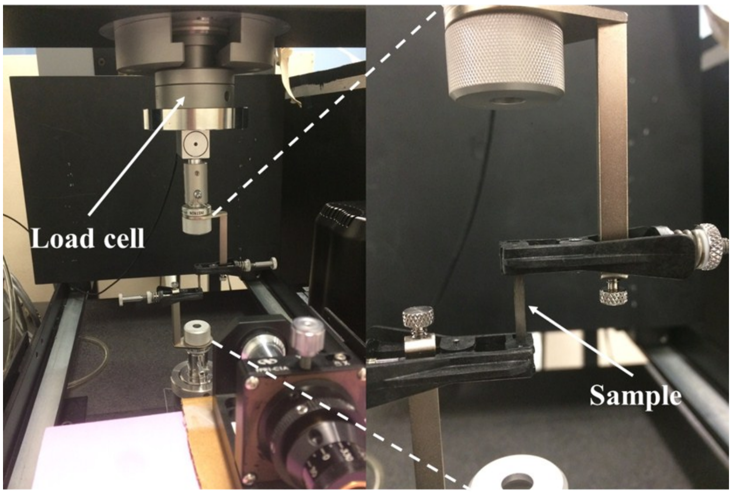

Fig. S5. Photograph of the experimental setup used for measuring photoinduced contraction force under the isostrain condition.

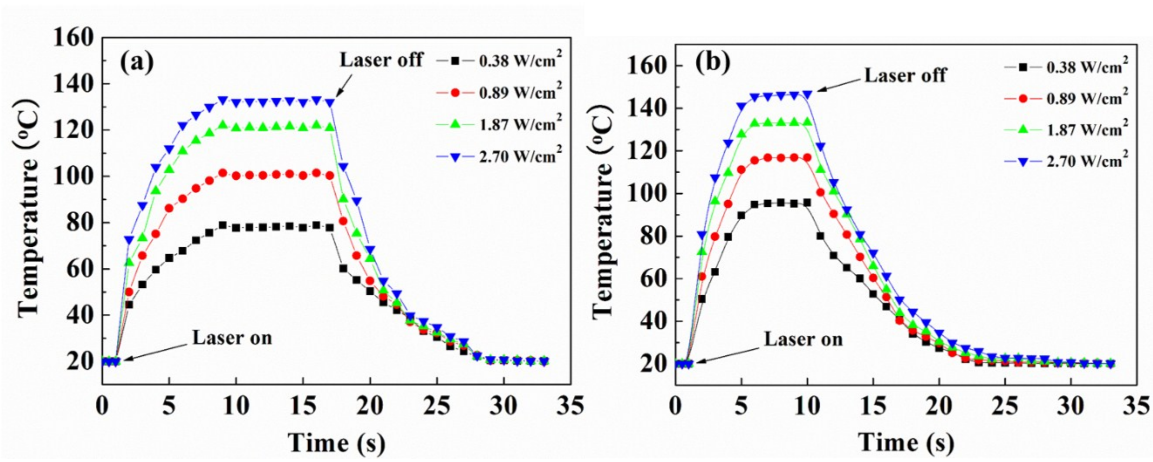

Fig. S6 Temperature changes of (a) actuator-3 and (b) RGO/ acrylic ester bilayer as a function of time upon NIR irradiations at different intensities. The NIR laser was applied to the RGO layer and the temperature on the other side of the bilayer or trilayer was measured using an infrared camera. In both cases, the strip was held straight to avoid delamination under prolonged NIR irradiation.

References

1) D. Li, M. Müller, S. Gilje, R. B. Kaner and G. G. Wallace, *Nat. Nanotechnol.*, 2008, **3**, 101.

Supporting Movie Captions

Video S1. Bending/unbending of an actuator in response to NIR laser (1.87 W/cm^2) on/off, respectively

Video S2. NIR light-driven wave propagation under NIR laser (1.87 W/cm^2) scan on a trilayer actuator strip whose two ends were fixed to the substrate.

Video S3. Caterpillar-like locomotion of a free actuator on untreated substrate surface (fluorescent plate) under NIR laser scan (1.87 W/cm^2).

Video S4. Caterpillar-like crawling up of a free actuator on untreated substrate surface (fluorescent plate) set at 15^o incline under NIR laser scan (1.87 W/cm^2).