# **Supporting Information**

## **Organic Liquid-Crystal Devices Based on Ionic Conductors**

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#### Movie S1

The liquid-crystal device is connected to a voltage source via two aluminum foils. A piece of paper with a logo is placed below the device, with the logo right beneath the active area of the device. At the voltage-off state, the active area is opaque and the logo is obscured. At the voltage-on state, the active area gradually becomes transparent and the logo is visible. After the voltage is switched off, the logo is concealed again. The movie is recorded using a digital camera (Canon, EOS 70D).

### Movies S2~S4, S6

The device is placed on the objective table of an optical microscope (Leica, DM 4000M), with one linear polarizer placed before the device, and without or with an analyzer after, in either perpendicular or parallel orientation. The central region of the active area is adjusted to be right under the objective lens. After tuning the focal length, voltage is applied through two aluminum foils and the dynamic behavior of the liquid crystals is recorded. Microscope images and videos are taken under three conditions: without analyzers, with analyzer in parallel orientation and with analyzer in perpendicular orientation. A cell phone is mounted onto the eyepiece to take the videos.

#### Movie S5

The device is mounted on a home-made biaxial stretcher which converts circular movement into radial movement. A voltage of amplitude 1200 V and frequency 1 kHz is applied to the device through two aluminum foils. Initially, the applied voltage is not high enough to switch on the device and the working area is opaque. Then the device is biaxially stretched, the thickness of the liquid crystal layer decreases and the electric field across the liquid crystal layer increases. When the stretch is larger than a critical value, the electric field is higher than the threshold value and the working area becomes

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transparent. The device becomes opaque again during the subsequent releasing process. The video is recorded using a digital camera (Canon, EOS 70D).



Fig. S1. Schematic of determining switch on time and switch off time.  $T_{saturated}$  is defined as the difference between the initial transmittance  $T_{initial}$  and the final transmittance  $T_{final}$ . The switch-on time  $\tau_{on}$  is defined as the time needed for the transmittance to increase from 10%  $T_{saturated}$  to 90%  $T_{saturated}$ . The switching-off time  $\tau_{off}$  is defined as the time needed for the transmittance to reduce from 90%  $T_{saturated}$  to 10%  $T_{saturated}$ .



Fig. S2. Electrooptical characteristics of a liquid-crystal device using ITO. (a) Schematic of the device architecture. (b) Transmittance of the device as a function of time under square-wave voltage of various amplitudes at a frequency of 1 kHz. (c) Switch-off time  $\tau_{off}$  is almost constant, ~8.2 ms in average, at various applied voltage. Note that here the thickness of liquid crystal layer should be larger than 20 µm even a pre-stretch of ~5 is applied to VHB, since the hole edge is a free boundary and some roughness of VHB is introduced during the fabrication process. When we applied voltage of amplitude up to 350 V on the device, we do not observed electric breakdown of the liquid crystals.



**Fig. S3. Image of the electrooptical device mounting on an equal-biaxial stretcher.** The stretcher converts circular movement into radial motion.



Fig. S4. Deflection of the dielectric membrane caused by the Maxwell stress.

We model each dielectric sheet as a pre-stretched membrane, anchored at the edge of the cell. When a voltage is applied on the two ionic conductors, the electric field causes a Maxwell stress in the dielectrics, pulling the two the dielectric sheets to deflect, and causing the liquid crystal to flow. Let the deflection of each membrane be w, the tension in the membrane be T (force per unit width), the pressure applied on the membrane be p. In this estimate, we neglect the effect of the ionic conductors. The deflection of the membrane is governed by the Poisson equations:

$$\frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} = \frac{p}{T}$$

For a membrane of radius R with a constrained boundary, the deflection is given by

$$w = \frac{1}{4} \frac{p}{T} \left( r^2 - R^2 \right),$$

where *r* is the radius of a point in the membrane, and *R* is the radius of the cell. We model the membrane as an incompressible, neo-Hookean material. In the undeformed state, the thickness of the membrane is *H*. When the membrane is under a stretch  $\lambda$ , the thickness becomes  $H\lambda^{-2}$ , and the tension is

$$T = \mu H \lambda^{-2} \left( \lambda^2 - \lambda^{-4} \right),$$

where  $\mu$  is the shear modulus. The Maxell stress is given by

$$p = \frac{1}{2} \varepsilon_{\rm o} \varepsilon E^2,$$

where  $\varepsilon$  is the dielectric constant of the membrane,  $\varepsilon_0$  is the permittivity of vacuum, and *E* is the electric field, related to the applied voltage *V* and thickness of the membrane as

$$E=\frac{V}{H\lambda^{-2}}.$$

A combination of the above expressions gives the deflection of the membrane:

$$w = \frac{1}{8} \frac{\mathcal{E}\mathcal{E}_{0}}{\mu} V^{2} \frac{1}{H^{3}} \frac{1}{\lambda^{-6} \left(\lambda^{2} - \lambda^{-4}\right)} \left(r^{2} - R^{2}\right)$$

The largest deflection occurs at the center of the membrane, r = 0.

In the following estimates, we use representative values,  $\varepsilon = 4.7$ ,  $\varepsilon_0 = 8.85 \times 10^{-12}$  F m<sup>-1</sup>,  $\mu = 100$  kPa,  $H = 500 \mu$ m, R = 1 cm, prestretch  $\lambda = 2$ . When V = 100 V, we estimate the deflection at r = 0 to be  $w \sim -6.8 \mu$ m. When applied voltage V = 1000 V, we estimate the deflection at r = 0 to be  $w \sim -680 \mu$ m, which will cause the two membranes to touch each other. The deflection under voltage will squeeze the liquid crystal from the middle to edge of the cell. The deflection can be removed by reducing the dimensions of the dielectric cell. For example, reducing the radius from 1 cm to 100  $\mu$ m decreases the deflection by four orders of magnitude.



**Fig. S5. Fabrication process of a liquid-crystal device.** (a) Cutting a circular hold in a dielectric using a laser cutter, followed by laminating another dielectric. This step produces a mold with a hole. The mold is then equal-biaxially stretched via a homemade stretcher. The thickness of the bottom layer dielectric and the hole are reduced, as indicated in the inset. (b) Liquid crystal is transferred into the hole and sealed by a top layer of dielectric. Ionic conductors are laminated and electronic conductors are placed on two sides respectively to form an ionic liquid crystal device. The dash circle indicates the opaque area containing the liquid crystal and the dash lines show the transparent areas covered with ionic conductors.



**Fig. S6. Experimental setup to observe microstructures in liquid crystal.** The liquid-crystal device is placed on the objective table of microscope, connecting to voltage source via two aluminum foils. The central region of the active area is adjusted to be right under the objective lens. After tuning the focal length, voltage is applied and the dynamic process of the liquid crystal realignment is recorded.