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journal or publication title	Physical Review E
volume	71
number	061702
page range	061702-1-061702-8
year	2005-06
URL	<a href="http://hdl.handle.net/10228/621">http://hdl.handle.net/10228/621</a>

doi: 10.1103/PhysRevE.71.061702

**Low-voltage-driven electromechanical effects of swollen liquid-crystal elastomers**

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(Received 13 April 2004; published 9 June 2005)

We experimentally investigate, in detail, electromechanical effects in liquid-crystal elastomers (LCEs) previously swollen with low-molecular-weight liquid crystals (LMWLCs). Both polydomain (POLY) and monodomain (MONO) LCEs were studied. We used a well known LMWLC, 4-*n*-pentyl-4-cyanobiphenyl (5CB) as a solvent. After swelling POLY and MONO LCEs (LSCE) with 5CB, shape changes were measured by recording the displacement of the edge of the swollen LCE at different voltages,  $V$ , and temperature. With 100  $\mu\text{m}$  distance between electrodes, measurable shape changes ( $\sim 1\text{--}20\ \mu\text{m}$ ) are observed with small voltages ( $V \sim 0.5\text{--}10\ \text{V}$ ). In particular, we note that, compared to unswollen L(S)CEs, a dramatic  $\sim 200$  times decrease of the threshold field was found for electromechanical effects in swollen L(S)CEs. While swollen MONO LCEs showed electromechanical effects in the planar geometry, homeotropic MONO swollen with homeotropically oriented 5CB did not. This is easy to understand because, in the homeotropic case, the liquid-crystal preferred axis is already aligned with the field so the field has no reorienting effect. The inverse of the response time when the field was switched *on* in both POLY and MONO was proportional to  $E^2$ , which is the same field dependence as the response time of LMWLCs. When the field was switched *off*, the relaxation time showed a field dependence different from that of LMWLCs that we attribute to relaxation of the LCE network.

DOI: 10.1103/PhysRevE.71.061702

PACS number(s): 61.30.Gd, 83.80.Va, 77.65.-j

**I. INTRODUCTION**

In the past three decades, swelling properties of isotropic polymer gels in isotropic solvents have attracted much attention and their physical mechanisms are now well understood [1]. On the other hand, a liquid-crystal elastomer (LCE), an anisotropic gel, has been recently synthesized, for which swelling properties in an anisotropic solvent such as low-molecular-weight liquid crystals (LMWLCs) are different

from the well-known isotropic case [2–4]. In particular, we found that the swelling properties and temperature behavior (thermomechanical property) of volume changes were also anisotropic [2–4].

Understanding the physical mechanisms responsible for thermomechanical effects in LCEs is important to make functional materials that can work as, e.g., temperature-driven actuators or artificial muscles [5–7]. Another useful property for them would be as devices that changed their shape in applied electric fields rather than temperature, i.e., their electromechanical effect. In current dry LCEs, however, very high fields are required to induce electromechanical effects [8].

In contrast to this, the electric field response of LMWLCs in the parallel plate geometry is a well-known cooperative

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phenomenon [9], i.e., is a voltage effect, not a field effect as it is for nematic elastomers. As a result, LCEs swollen with nematic LMWLCs could be a good candidate to observe measurable shape changes at low voltages. Indeed, only a small voltage ( $\sim 1$  V) is required to change the orientation of nematic LMWLCs. Should there be a strong enough coupling between the mesogenic sidechains of the LCE and LMWLC molecules, the question is can the low voltage response of LMWLCs be harnessed to induce reorientation effects in the tethered LCE side chains to obtain measurable shape changes at low voltages? Our brief answer is yes. However, while the voltages are small, comparable to that of LMWLCs, the reorientation effect is also small.

In the first investigation of electromechanical effects in LCEs swollen with LMWLCs in 1986, Zentel found [10] tiny changes in large fields. Subsequently, Barnes *et al.* [11] reported their largest field driven shape change to be about 20% contraction of polydomain elastomer swollen in 6CB (hexyl-cyano-hexyl-biphenyl) when both, the elastomers and 6CB, were isotropic. In our study, we find no electromechanical effect when 5CB is isotropic.

In 1994, Kishi *et al.* [12] reported quantitative results on shape changes of swollen polydomain LCEs under a dc electric field ( $0.3$  V/ $\mu\text{m}$ ). Kishi *et al.*'s geometry has similarities to our geometry but in the elastomer/LMWLC that we study, a maximum 13% contraction is observed in fields ranging from  $0.01$ – $1$  V/ $\mu\text{m}$ .

Chang *et al.* [13] studied electro-optic effects in nematic gels obtained by photo-crosslinking a well-oriented LMWLC. They found that relatively high voltages were required to observe a small director reorientation. For our purposes, the relevant feature of this paper is that they were able to account for all their effects by invoking only LMWLC elasticity with the network effects taken care of by a nearly constant field ( $E_0 = V_0/d \sim 0.11$  V/ $\mu\text{m}$  with sample thickness  $d$ ) applied perpendicular to the applied field ( $\sim 0.3$  V/ $\mu\text{m}$ ). To obtain fits to their data, they had to assume a characteristic length scale of  $\sim 1$   $\mu\text{m}$  for their gels, which were homogeneous samples,  $62$   $\mu\text{m}$  and  $125$   $\mu\text{m}$  with a  $3^\circ$  pretilt (the director is not exactly perpendicular to the field direction before the field is applied). As is well known, in a parallel-plate geometry, well-oriented nematic LMWLCs have no length scale. As a result, electric field reorientation effects, not depending on sample thickness (to first order), are a voltage effect not a field effect.

Recently, Courty *et al.* [14,15] demonstrated a fairly large electromechanical response in an LCE embedded with a low concentration of carbon nanotubes. They needed a large applied field (order of  $\sim 1$  V/ $\mu\text{m}$ ) to generate a strain,  $\epsilon$ , of order  $\sim 0.1\%$ . These results raise the possibility that, if long range orientational order of elastomer sidechains are "strong" enough to reorient nonmesogenic rodlike objects, there could be an even stronger and lower voltage effect between LMWLCs "embedded" in a swollen LCE and the LCE sidechains also with long range orientational order.

Here, we investigate electromechanical effects in monodomains and polydomains of specific liquid crystalline elastomers after they have been swollen with a well-known low-molecular-weight liquid crystal. Two important observations from our previous swelling study [2–4] of this special system

are (1) 5CB enters the bulk of the LCEs (i.e., is not confined to the LCE-5CB interface) where it aligns with the LCE sidechains and (2) the LSCE length parallel to the director does not change during swelling. In Sec. II, we describe the sample preparation and experimental setup. The statics and dynamics of the observed shape changes are discussed in Sec. III, e.g., only a small voltage ( $V \sim 0.5$ – $10$  V) is required to obtain a measurable ( $\sim 1$ – $20$   $\mu\text{m}$ ) shape change and it took about 10 s to reach the maximum shape change ( $20$   $\mu\text{m}$ , i.e., comparable to half the swollen elastomer thickness). Section IV contains a summary and some perspectives.

## II. THE EXPERIMENT

### A. LCE sample preparation

The LCE materials used here were invented and synthesized by Finkelmann and co-workers [16–19]. The structure of LCEs involves mesogenic groups chemically tethered as side chains to a cross-linked polymer network [16–18]. The ingredients and procedure of the LCE synthesis have been described in our previous paper [4].

Polydomain LCEs are produced in which there are domains with different director orientations, denoted by a unit pseudovector,  $\hat{\mathbf{n}}$  [17]. But then it was discovered that applying a large enough strain could reorient the domains giving rise to a uniform director orientation [17,19]. Then monodomain samples, i.e., liquid single crystal elastomers (LSCEs), were successfully made using two cross-linking steps with the second cross-linking step performed on a stretched film [18]. These films turned out to have a spatially uniform  $\hat{\mathbf{n}}$  parallel to the stretching direction,  $\hat{\mathbf{x}}$  in Figs. 1.

Figure 1(b) is a snapshot in the  $\hat{\mathbf{x}}$ - $\hat{\mathbf{y}}$  plane of the swollen LSCE sample in zero field. Its sample dimensions are  $l_y = 270$   $\mu\text{m}$  and  $l_z = 45$   $\mu\text{m}$ .  $l_x = 300$   $\mu\text{m}$  was the same before and after swelling. During swelling, 5CB invades the LSCE about three times faster  $\parallel \hat{\mathbf{n}}$  than  $\perp \hat{\mathbf{n}}$  [3,4].

In Fig. 1(c), the director field is sketched for each of the two types of interfaces. The gray scale bar above each sketch depicts the optical contrast observed looking "down" on the sample ( $\hat{\mathbf{x}}$ - $\hat{\mathbf{y}}$  plane). In the LSCE, the director configuration is taken to be antisymmetric. This is in contrast to the usual case for a Fréedericksz transition in a parallel plate geometry. A consequence of antisymmetric boundary conditions is that the LMWLC director at the center of the LMWLC has two competing effects for reorientation so is constrained to be parallel to  $\hat{\mathbf{x}}$  even in a very large field at the LCE center.

To match the boundary conditions at the LSCE-5CB interfaces, there must be several  $S = \pm 1/2$  line defects [e.g., the dotted line marked "a" in the bend-splay schematic of Fig. 1(c) and the thick black line in Fig. 1(b)] some of which will move in a field aligning  $\hat{\mathbf{n}} \parallel \mathbf{E}$ .

Shape changes are measured by observing the displacement of the bend-splay interface between the swollen LSCE and the embedding LMWLC ( $\Delta l_x = \delta$ ) with applied voltage. Only a small effect is observed at the twist interfaces as the director remains always (and only) in the  $\hat{\mathbf{x}}$ - $\hat{\mathbf{z}}$  plane. In other words,  $n_y \sim 0$  everywhere and the whole sample looks dark when polarizer and analyzer are crossed parallel to  $\hat{\mathbf{n}}$ .

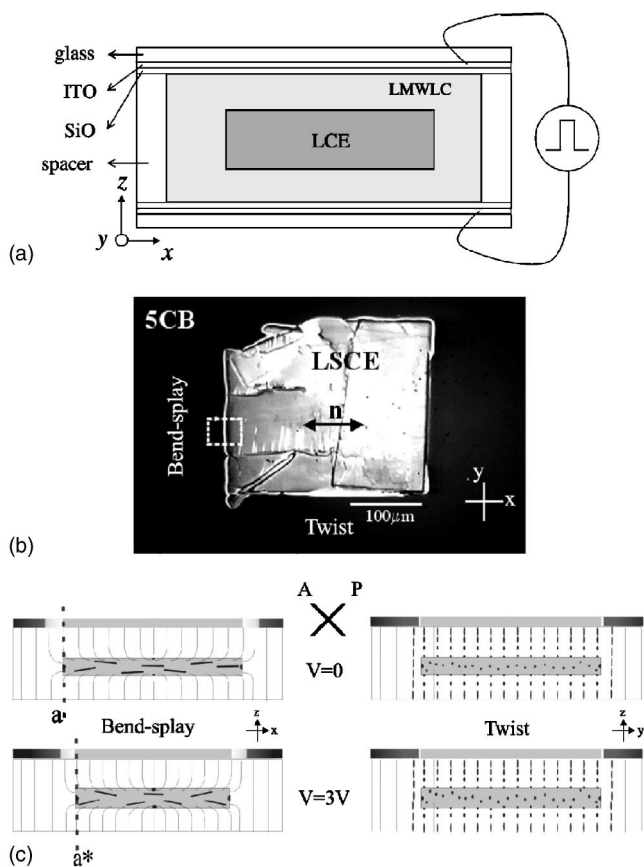


FIG. 1. (a) Experimental setup. An alternating electric field,  $\mathbf{E} = (0, 0, E_z)$ , is applied to the sample cell. The distance between the ITO electrodes is  $100 \mu\text{m}$ . The LCE is embedded in LMWLC (5CB). While 5CB is oriented  $\parallel \hat{z}$  (homeotropic) at the electrodes, it rotates  $\pi/2$  in a splay-bend mode to be  $\parallel \hat{x}$  inside the elastomer. (b) Snapshot, crossed polarizers oriented  $45^\circ$  to the picture edges, of the swollen LSCE where the director,  $\hat{\mathbf{n}}$ , is  $\parallel \hat{x}$ . The two kinds of interfaces between the homeotropically oriented LMWLC (black regions) and the LSCE (gray region) are (1) the bend-splay interfaces  $\parallel \hat{x}$  and (2) twist interfaces  $\parallel \hat{y}$ . We interpret the nearly vertical dark line running through the LSCE in (b) as an  $S = +1/2$  line defect implying an antisymmetric configuration for 5CB in the elastomer which suppresses the usual Fréedericksz transition in LMWLCs. Measurements are made in the middle of the bend-splay interface shown as a dotted square. (c) Director schematics of the interfaces with gray scale above each showing the optical contrast between crossed polarizers set  $45^\circ$  to  $\hat{\mathbf{n}}$  in the  $\hat{x}$ - $\hat{y}$  plane. The dotted line marked  $a$  denotes the  $S = -1/2$  disclination line that we track and which moves to  $a^*$  when 3 V is applied as discussed in the text.

The polydomain (POLY), planar monodomain (P-MONO) and homeotropic monodomain (H-MONO) LCE films we prepare are  $\sim (25.0 \pm 1.0) \mu\text{m}$  thick with area  $\sim 300 \mu\text{m} \times 150 \mu\text{m}$ . P-MONO is obtained by slicing the LSCE parallel to  $\hat{\mathbf{n}}$  and H-MONO is obtained by slicing perpendicular to  $\hat{\mathbf{n}}$  [4]. The samples are embedded in LMWLCs (anisotropic solvents) for swelling between two transparent indium tin oxide (ITO) electrodes with very clean SiO surfaces as shown in Fig. 1(a). The SiO coating is to avoid complications arising from charge injection. In this study we use the well-known nematic LMWLC, 4-n-pentyl-4-cyanobiphenyl

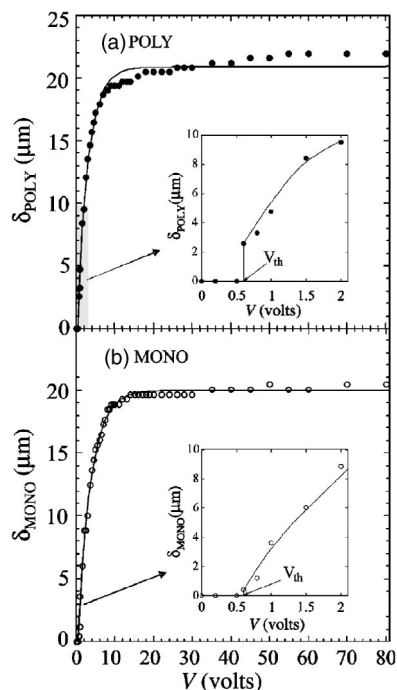


FIG. 2. Voltage dependence of the displacement  $\delta$ ,  $T = 26^\circ\text{C}$ . The distance between electrodes is  $100 \mu\text{m}$ . (a) In POLY,  $\delta_{POLY} = (20.87 \pm 0.16) - (22.38 \pm 0.43)\exp(-V/2.88 \pm 0.17)$ . (b) In planar (P-) MONO,  $\delta_{MONO} = (20.00 \pm 0.12) - (22.12 \pm 0.32)\exp(-V/3.38 \pm 0.10)$ . Insets: expanded version of the graph to show the threshold for the LCE shape change;  $V_{th}^{POLY} \sim 0.6 \text{ V}$  for POLY (a) and  $V_{th}^{P-MONO} \sim 0.6 \text{ V}$  (b).

(5CB) that was homeotropically aligned [Fig. 1(b)] at the electrode surface. The cell gap was controlled by a  $100 \mu\text{m}$  polymer (Mylar) spacer.

In the experiments, an alternating electric field of frequency  $f = 50 \text{ Hz}$  with rectangular waveform is applied between the electrodes, i.e.,  $\mathbf{E} = (0, 0, E_z)$  at constant temperature  $T = 26^\circ\text{C}$  controlled to  $\pm 0.1^\circ\text{C}$  by a temperature regulator (CHINO Digital Controller DB500).

**B. Electromechanical effect measurement**

Measurements of the electromechanical effect were made using a polarizing optical microscope (Nikon E600WPOL) on which a charge-coupled-device camera (SONY XC-75) was mounted. For monodomain LCE (LSCE), the director  $\hat{\mathbf{n}}$  was set parallel to the  $\hat{x}$  direction. The mechanical deformation and temporal changes of the electromechanical effect were captured by a computer. Here, we used an image capture software (Scion Image). Shape changes are measured by observing the displacement of the bend-splay interface between the swollen LCE and the embedding LMWLC ( $\Delta l_x = \delta$ ) with applied voltage [Fig. 1(b), Fig. 2].

**III. MEASUREMENTS AND DISCUSSIONS**

**A. Low-voltage-driven shape changes**

As described in our previous communications [3], the equilibrium swollen state of L(S)CEs swollen with 5CB is

achieved after about 2 h. After swelling, the dimensions of dry P-MONO perpendicular to  $\hat{n}$ ,  $l_y \sim 150 \mu\text{m}$  and  $l_z \sim 25 \mu\text{m}$  change to  $\sim 270 \mu\text{m}$  and  $\sim 45 \mu\text{m}$  respectively, while the dimension parallel to  $\hat{n}$ ,  $l_x$ , does not change. The POLY sample swells isotropically in all three dimensions by a factor of about 1.8.

In Fig. 2, the solid lines are fits to

$$\delta_{POLY}(V) = 20.87 - 22.38 \exp(-V/2.9), \quad (1)$$

for the swollen POLY and

$$\delta_{MONO}(V) = 20.00 - 22.12 \exp(-V/3.4), \quad (2)$$

for swollen P-MONO, as a ‘‘guide to the eye.’’

The insets in Fig. 2 show relatively small threshold values,  $V_{th}$ , for the onset of the observed electromechanical effect:  $V_{th}^{POLY} \sim 0.6\text{V}$  for the swollen POLY sample [Fig. 2(a)] and  $V_{th}^{P-MONO} \sim 0.6\text{V}$  for the swollen planar P-MONO sample [Fig. 2(b)]. Note also that while reorientation at the threshold is sharp for POLY (suggesting a subcritical bifurcation), it is quite smooth for P-MONO. That is, the bifurcation here is imperfect, which is the usual case for Fréedericksz transitions when the director is not exactly perpendicular to the applied field (pretilt).

To compare this to dry LCEs where director reorientations are a field effect, we need to convert volts to field. In dry LCE, the displacement is observed at about  $\sim 1\text{V}/\mu\text{m}$  [8,14]. Therefore, by assuming that the threshold field for swollen L(S)CE is about  $0.5\text{V}/100\mu\text{m}$  (cell thickness is  $100\mu\text{m}$ ), it is about 200 times smaller than that of dry LCEs.

Figure 3 shows schematic pictures for the mechanism of the electromechanical effects of the swollen LCE. Applying an electric field to an LCE swollen with LMWLC, when the LMWLCs director reorients parallel to the field. This induces the reorientation of many mesogenic sidechains that in turn change the network shape making the sample thicker along  $\mathbf{E}$  and thinner perpendicular to the bend-splay interface  $\mathbf{E}$  and very little (if at all) on the twist interface (because  $n_y \equiv 0$  independent of  $\mathbf{E}$ ).

Thus the electromechanical effect is induced by the cooperative effect with LMWLC. The mesogenic side chains in swollen LCEs can effectively couple with each other via LMWLC molecules dispersed in networks. Therefore a relatively weak electric field triggers a macroscopic shape change. Dividing the maximum displacement ( $\sim 40\mu\text{m}$ , i.e.,  $\sim 20\mu\text{m}$  at each interface) by the zero field sample length ( $\sim 300\mu\text{m}$ ), contractions of about 13–14% are found (e.g., see Fig. 4 for swollen P-MONO).

Figures 4 are optical micrographs at large magnification of P-MONO without a field ( $V=0$ ) and in a field ( $V=3\text{V}$ ). Homeotropically oriented 5CB is black for polarizers crossed at any orientation. To obtain extinction in well-oriented homogeneous samples, the polarizer and analyzer must be crossed parallel (or perpendicular) to  $\hat{n}$ . Maximum intensity is achieved when they are crossed  $45^\circ$  to this special direction. The very bright region between the elastomer and the LSCE is where 5CB starts to reorient from being  $\parallel \hat{z}$  far from the Bend-splay interface to be  $\parallel \hat{x}$  inside the elastomer. This bright region is a result of 5CB’s increasing birefringence as

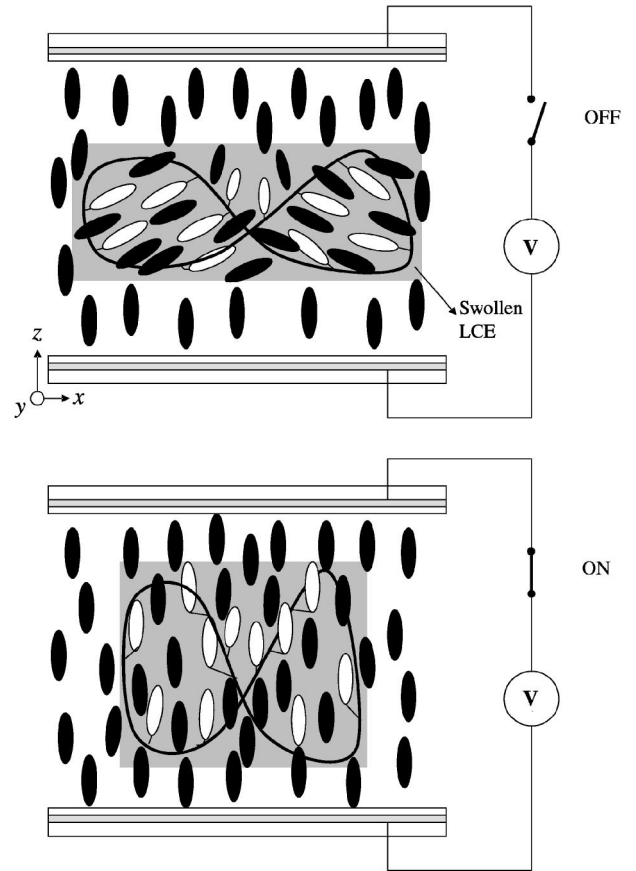


FIG. 3. Schematic picture for the electromechanical effect of LCE swollen with LMWLC without a voltage (top) and with (bottom). On the bottom, the LCE network has been stretched in the direction of  $\mathbf{E}$  and shrunk perpendicular to  $\mathbf{E}$ . Here, the dark and open ellipses indicate LMWLC and mesogenic side chain molecules, respectively. The line indicates the polymer network.

its extraordinary optic axis rotates to meet that of the SLCE  $\parallel \hat{x}$ . Loosely speaking, the sample optically thickens on going from homeotropic 5CB to the LSCE similar to a lens in isotropic systems.

There are only singular solutions for a nematic director to rotate  $\pi/2$  in a finite distance. Hence, the need for  $S = \pm 1/2$  defect lines [Figs. 1(b) and 1(c)]. At the line marked  $a$ , the birefringence abruptly increases. This we attribute to a defect line at the elastomer edge in contact with 5CB. The line marked  $b$  is the double image of the defect line nearly in the same focal plane as  $a$  when  $V=0$ .

The distance between  $a$  and  $b$  is about  $15\mu\text{m}$ . Applying a voltage of  $\sim 3\text{V}$ , this distance shrinks by about  $10\mu\text{m}$ ,  $a$  shifts to  $a^*$  ( $\sim 10\mu\text{m}$  to the right of its  $V=0$  position) and  $b$  shifts to  $b^*$  ( $\sim 1\mu\text{m}$ ) and thickens as it moves out of the focal plane of  $a^*$ . When the field is turned on (3 V),  $a$  moves as the 5CB birefringent region narrows so the defect double image is now focussed above the focal plane of  $a^*$ .

In Fig. 5, both LMWLC and LSCE have been prepared homeotropically. In the equilibrium swollen state, we pointed out that the LMWLC molecules should be parallel to the LSCE sidechains inside the interfacial region. When an electric field is applied, no electromechanical effect can be ob-

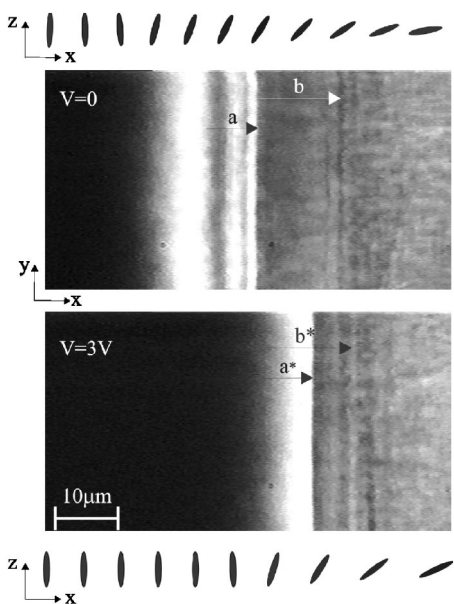


FIG. 4. Large magnification optical micrographs of P-MONO's bend-splay interface with (bottom) and without (top) an applied voltage. Average director schematics are shown for each picture above the picture ( $V=0$ ) and below for  $V=3$  V. Polarizer and analyzer crossed  $45^\circ$  to the micrograph edges and also  $\hat{n} \parallel \hat{x}$  of the LSCE.

served because there is no reorientation of the LMWLC molecules and the mesogenic sidechains inside the interfacial region [Figs. 5(a) and 5(b)].

**B. Question: Bulk or surface effect?**

In swollen P-MONO, field-induced shape changes are observed in the vicinity of the swollen LCE-LMWLC interface

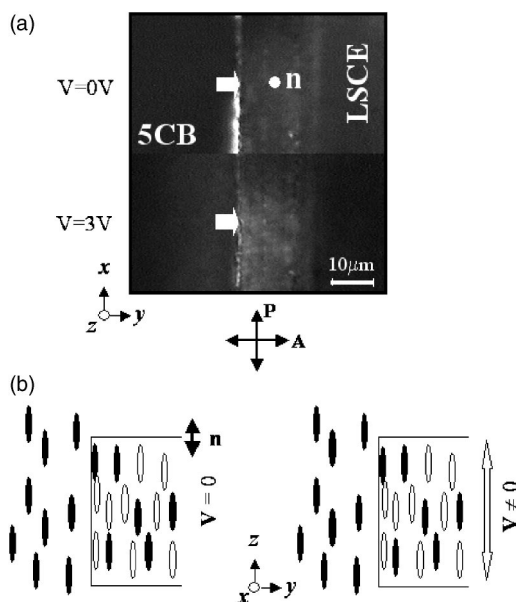


FIG. 5. (a) Optical micrographs in crossed polarizers showing that there is no electromechanical effect when both the LSCE and the LMWLC are homeotropically oriented, H-MONO sample. (b) Schematic of (a). The LMWLCs are represented by dark ellipses.

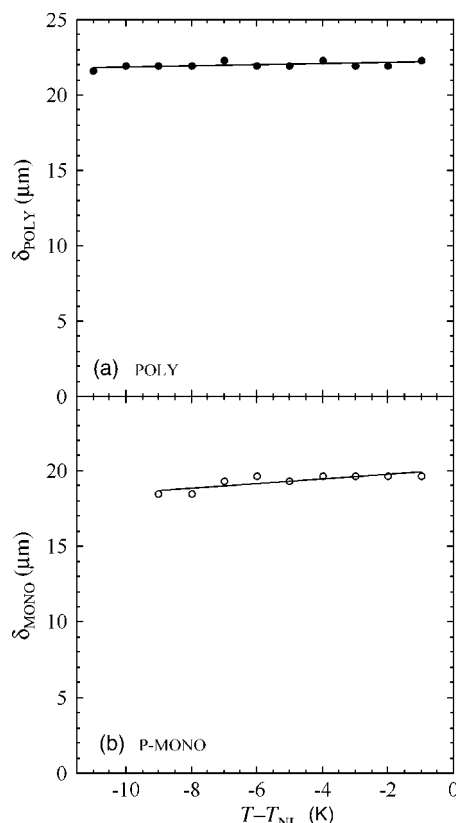


FIG. 6. Temperature dependence of the displacement  $\delta$  in a field ( $V=50$  V). (a) In POLY,  $\delta_{POLY}=22.21+0.03(T-T_{NI})$ . (b) In P-MONO,  $\delta_{MONO}=20.09+0.15(T-T_{NI})$ .

where the director of the LMWLC changes its orientation from being  $\parallel \hat{z}$  to  $\parallel \hat{x}$ . While the electromechanical response we have observed so far is measurable and reproducible, we would like to understand better how to make it larger.

One reason limiting the effect could be the smecticlike properties of this SLCE [3] constraining the 5CB inside the elastomer. Another could be constraints on 5CB because of the antisymmetric boundary conditions of the geometry as depicted in Fig. 1(c) and evidenced by the presence of the  $S=+1/2$  wrapped around the elastomer in the  $\hat{y}-\hat{z}$  plane [the thick vertical line referred to in Fig. 1(b)].

For applications requiring a larger effect, then, we need more experiments with different LCEs and different geometries.

**C. Temperature dependence of shape changes**

Figure 6 shows the temperature dependence of the displacement  $\delta$  in an electric field ( $f=50$  Hz and  $V=50$  V). In the swollen POLY, as temperature  $T$  is increased, the displacement increases very slightly,  $\delta_{POLY}=22.21+0.03(T-T_{NI})$  [Fig. 6(a)], where  $T_{NI}$  is the phase transition temperature for the nematic-isotropic transition in 5CB. In the swollen P-MONO, the displacement also increases slightly with  $\delta_{MONO}(T)=20.09+0.15(T-T_{NI})$  [Fig. 6(b)]. In these two systems, a maximum relative contraction is achieved at a temperature just before the nematic-isotropic phase transition

temperature  $T_{NI}$ . In the isotropic phase, no displacements are observed.

As a reorientation of the LMWLC molecules does not occur when the LMWLC is isotropic, it cannot drive a shape change. In our previous papers [2–4], we have found that the apparent nematic-isotropic phase transition temperature of the swollen LCEs is  $\sim 80^\circ\text{C}$  for POLY and MONO. When 5CB is in its nematic phase, the contraction of swollen L(S)CE in a low electric field ( $E \sim 0.5\text{ V}/\mu\text{m}$ ) is about  $20\ \mu\text{m}$  at maximum. While 5CB goes isotropic at  $T_{NI} \sim 34.5^\circ\text{C}$ , the liquid crystalline parts of the LCE network are still in their nematic phase until  $\sim 80^\circ\text{C}$  for POLY and MONO. In this case it is still possible to have a small contraction, but the application of a high electric field is necessary.

In summary it can be stated that electromechanical effects observed in swollen L(S)CEs are a consequence of the cooperative effects of the LMWLC embedded in the swollen L(S)CE with the sidechains and the network of the LCE. However, the effect is much smaller than expected. We have suggested that this could be because of the boundary constraints on homeotropically oriented 5CB and the LCE interfaces and also the smecticlike properties of the elastomer “infecting” 5CB. This can be tested by redoing the experiment with homogeneously oriented 5CB, which would minimize both the necessity for defect lines and reduce the possibility of antisymmetric boundary conditions at the LSCE-5CB interfaces. Another test would be to redo the experiment with another LSCE with less pronounced smecticlike features [3].

#### D. Space-time graphs and response times

To quantify the shape change dynamics of the swollen L(S)CE in an applied electric field, we analyzed the data obtained from space-time plots.

Figures 7 and 8 show the voltage dependence of the normalized displacement  $\delta_{nlz}$  in the shrinking (switched *on*) and the stretching (switched *off*) processes, respectively, at fixed temperature  $T=26^\circ\text{C}$  and frequency  $f=50\text{ Hz}$ . The dotted lines in these figures are best fits to  $\delta_{nlz}(t, V) = \delta_0 \exp(-t/\tau_{on})$  and  $\delta_{nlz}(t, V) = \delta_\infty + \delta_0 \exp(-t/\tau_{off})$  for switching *on* and *off* the external field, respectively. It means that only a single process plays a role for the response, which may be due to reorientation dynamics of  $\hat{n}$ .

In Fig. 7, with increasing applied voltage,  $\delta_{nlz}$  in both swollen POLY and P-MONO samples decreases more rapidly.

In Fig. 8, when the voltage is switched *off*,  $\delta_{nlz}$  in the swollen POLY increases rapidly when the initial voltage is below 8 V, and there is almost no change above 8 V [Fig. 8(a)]. In swollen P-MONO, there was a relatively small change of  $\delta_{nlz}$  when the field is switched *off* [Fig. 8(b)].

Figures 9 and 10 show the voltage dependence of the response times for the swollen POLY and P-MONO samples, when the field is switched *on* and *off*, respectively.

In Fig. 9, the inverse of the response times ( $1/\tau_{on}$ ) in swollen POLY and P-MONO are clearly proportional to  $V^2$ . In the swollen POLY, the solid line is a fit to  $1/\tau_{on}^{POLY} = 0.51 + 0.10\text{ V}^2$  [Fig. 9(a)].

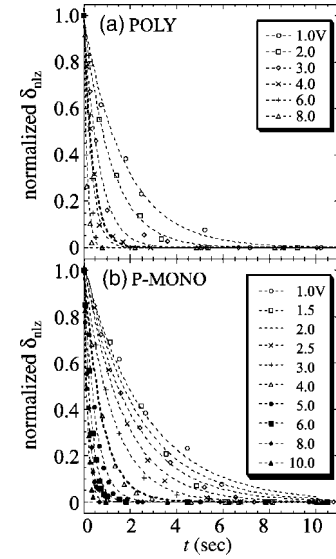


FIG. 7. Voltage dependence of the normalized displacement  $\delta_{nlz}$  in the shrinking (switching *on*) process for swollen POLY (a) and P-MONO (b) samples. The dotted lines are best fits to  $\delta_{nlz}(t, V) = \delta_0 \exp(-t/\tau_{on})$ .

In the swollen P-MONO, the solid line is a fit to  $1/\tau_{on}^{P-MONO} = 0.34 + 0.06\text{ V}^2$  [Fig. 9(b)]. This result is consistent with the response behavior of ordinary liquid crystals. In the swollen P-MONO sample, the response time for the shape changes is slightly higher than that in the swollen POLY samples. This effect could be due to the fact that the rotation of the LMWLC molecules and the mesogenic sidechains is resisted by the elastic network, an

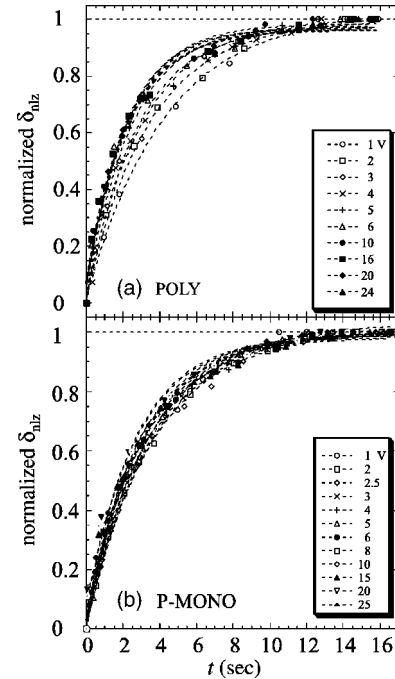


FIG. 8. Voltage dependence of the normalized displacement  $\delta_{nlz}$  in the stretching (switching *off*) process for swollen POLY (a) and P-MONO (b) samples. The dotted lines are best fits to  $\delta_{nlz}(t, V) = \delta_\infty + \delta_0 \exp(-t/\tau_{off})$ .

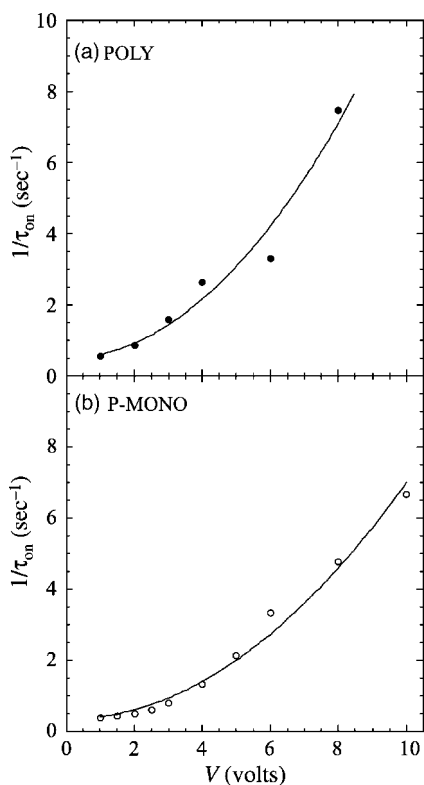


FIG. 9. Voltage dependence of the inverse response time of swollen POLY (a) and P-MONO (b) samples when the field is switched on.  $T=26^\circ\text{C}$ .

effect that is larger in P-MONO due to the stretching process during the LSCE preparation that endows the network with smectic features.

Figure 10 shows the inverse of the relaxation times ( $1/\tau_{off}$ ) for swollen POLY and P-MONO samples. In the swollen POLY, at first  $1/\tau_{off}^{POLY}$  increases with increasing applied voltage (until  $\sim 8\text{ V}$ ), and then goes to a constant ( $\sim 0.45$ ) above  $8\text{ V}$  [Fig. 10(a)]. In the swollen P-MONO, a

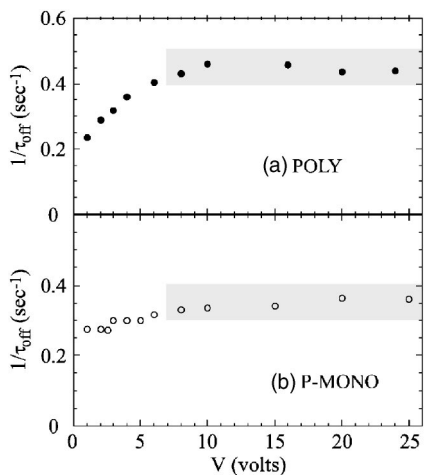


FIG. 10. Voltage dependence of the inverse relaxation time of the swollen POLY (a) and P-MONO (b) samples when the field is switched off and the electrodes shorted.  $T=26^\circ\text{C}$ . In the shaded regions above  $8\text{ V}$  the relaxation time is constant.

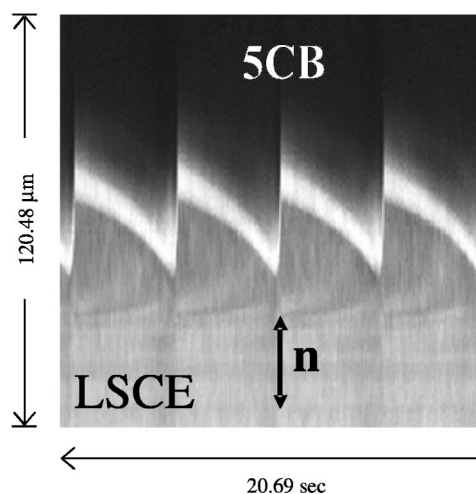


FIG. 11. Oscillations shown by a swollen LSCE at low frequencies ( $0.2\text{ Hz}$ ) displayed as a space-time plot.  $T=26^\circ\text{C}$  and  $V=10\text{ V}$ .

constant value of  $1/\tau_{off}^{P-MONO}$  ( $\sim 0.35$ ) is also observed with increasing voltage [Fig. 10(b)]. Below  $8\text{ V}$ , the inverse of the relaxation time  $1/\tau_{off}^{P-MONO}$  increases in voltage.

In ordinary liquid crystals, the relaxation time is independent of the initial voltage when the field is switched off. However, in swollen L(S)CEs, in the present study we found that this behavior is valid only above  $\sim 8\text{ V}$ ; below  $8\text{ V}$  it has a pronounced voltage dependence. We suggest that the behavior observed below  $8\text{ V}$  is due to the electromechanical energy stored in the L(S)CE network in the interfacial region and also in the bulk of swollen L(S)CE.

As one might expect from the results given in Figs. 9 and 10, a dynamical response can be observed in the form of periodic oscillations of shape changes in swollen MONO (LSCE) samples, when a sufficiently low frequency ( $f < 1/\tau_{off}^{P-MONO}$ ) is applied. An example is shown in Fig. 11 at  $T=26^\circ\text{C}$  and  $V=10\text{ V}$ .

IV. SUMMARY

We studied the electromechanical effects of L(S)CEs swollen with LMWLC and found the following main results.

The threshold field for electromechanical effects in swollen L(S)CEs is comparable to that of LMWLC and thus about two orders of magnitude smaller than that of dry L(S)CEs. The inverse response times after switching on the external field for both swollen POLY and MONO (LSCE) samples show a quadratic dependence on the field strength. This is consistent with the inverse response time for ordinary low molecular weight liquid crystals. From this observation we conclude that the liquid crystal features dominate the response of swollen L(S)CEs when the field is switched on. When the field is switched off, the inverse of the relaxation time is independent of voltage above  $8\text{ V}$  and depends on voltage below  $8\text{ V}$  for a sample thickness of  $\sim 100\ \mu\text{m}$ . This pronounced voltage dependence can be interpreted as being due to the electromechanical energy stored in the L(S)CE network.



## ACKNOWLEDGMENTS

We thank Nicole Assfalg for sending us some of the samples used in these measurements. P.E.C. thanks the Japan Society for the Promotion of Science for financial assistance that enabled this collaboration. This work is partially supported by the Japan-Germany Scientific Cooperative Pro-

gram of the Japan Society for the Promotion of Science and the Deutsche Forschungsgemeinschaft, and the Grant for Scientific Research sponsored by the Japan Society for the Promotion of Science. H.R.B. thanks the Deutsche Forschungsgemeinschaft for partial support of his work through Sonderforschungsbereich 481 “Komplexe Polymer- und Hybridmaterialien in inneren und äußeren Feldern.”

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