## Appendix A

## Transient Electro-Optic Properties of Liquid Crystal Gels

The dynamics of the electro-optic response has significance for the use of our gels in display devices and also reveals important effects of the polymer network. In order to study this behavior, we recorded the transmitted intensity before, during, and after applying a voltage to the gel. This allowed us to measure the response time for aligning the gel in the presence of a voltage and the relaxation time after the voltage was removed. In the following we analyze the dynamics of 5 wt % and 10 wt % gels confined to a 15  $\mu$ m gap. These samples reveal the general characteristics of the dynamics of the electro-optic response and its dependence on the applied voltage and the electro-mechanical history.

Cells were prepared with 25 mm x 25 mm x 1.1 mm Delta ITO-coated glass plates. The glass plates are  $SiO_2$  passivated and contain no alignment layers. The plates were fastened together using epoxy, and teflon spacers were placed in between the plates to create a well-defined gap. We measured the gap thickness with a micrometer, and loaded the cell by placing a small amount of gel on the side of the gap, heating it to 40  $^{\circ}$ C, and letting the gel flow in by capillary action.

The optical properties and electro-optic responses of the gels were measured using a polarized HeNe laser, a beam splitter, two CCD cameras, and a function generator. A beam splitter between the laser and the sample sent half of the incident laser intensity to the sample and half to a CCD camera which was used to normalize the intensity of laser light incident on the sample. A CCD camera 10 cm behind the sample measured the intensity of transmitted light, and this intensity was



Figure A.1: Response of a 5 wt % gel in a  $15\mu$  gap to an external voltage of 30 V, 60 V, and 90 V. The voltage is applied at 0 s and removed at approximately 15 s.

normalized by the intensity transmitted for the sample in the isotropic state. The function generator was used to apply voltages from 0 V to 270 V at 1000 Hz.

For each run, a voltage was applied for only 15 s. After allowing the gel to sit for 2 minutes at 0 V, the process was repeated with the same voltage 4 times, for a total of 5 runs for each experiment. Then, before the start of the next experiment, the gel was heated into the isotropic state and allowed to cool.

The response of the gels depends on the applied voltage (Fig. A.1). At low voltages, near the voltage threshold, the long-time transmission is very low, as would be expected. Voltages at twice and three times the threshold give a much higher long-time transmission. At 30V, the response is very slow but relaxation is fast. At higher voltages, the response time gets faster and the relaxation time slower. We can define a response time and relaxation time for the gel. The time for the gel to reach 90% of its long-time transmission after turning on the signal,  $\tau_{90}$ , is a characteristic response time for the gel. The time for the transmission to decay back down to 10% of its long-time transmission,  $\tau_{10}$ , is a characteristic relaxation time. The  $\tau_{90}$  are 10.0547 s, 3.7630 s, and 0.1070 s for 30 V, 60 V, and 90 V, respectively, and the  $\tau_{10}$  are 0.0160 s, 0.2508 s, and 0.1342 s for 30 V, 60 V, and 90 V, respectively, for the 5 wt % gel. This indicates that applying a high enough voltage imprints alignment into the polymer network, resulting in a slow response (larger  $\tau_{10}$ ). Also, at high

Polymer Concentration	Voltage	$ au_{90}  ({ m s})$	$\tau_{10}   (s)$
5  wt  %	$V_{threshold}(30V)$	10.0547	0.0160
5  wt  %	$2 \times V_{threshold}(60V)$	3.7630	0.2508
5 wt %	$3 \times V_{threshold}(90V)$	0.1070	0.1342
10 wt $\%$	$V_{threshold}(45V)$	8.1635	0.0007
10 wt $%$	$2 \times V_{threshold}(90V)$	4.4864	0.5533
10 wt %	$3 \times V_{threshold}(135V)$	0.1395	0.4510

Table A.1: Results for transient switching experiments of 5 wt % and 10 wt % gels in a 15  $\mu$  gap.

enough voltages the electric field dominates the dynamics and the liquid crystal director rotates very quickly, as indicated by a small  $\tau_{90}$  for large applied voltages.

We can study the effect of polymer concentration by comparing 5 wt % and 10 wt % gels of the same thickness (Tab. A.1). Both behave the same qualitatively, with a faster switch-on time and slower switch-off time at higher voltages. At low voltages, the 10 wt % gel has a much faster switch-off time than the 5 wt % gel – the  $\tau_{10}$  is 0.016 s for the 5 wt % gel compared with 0.0007 s for the 10 wt % gel. At twice the threshold voltage, the  $\tau_{90}$  values are comparable at ~ 4 s, but the  $\tau_{10}$  is 0.25 s for the 5 wt % gel compared to 0.55 s for the 10 wt % gel. At three times the threshold voltage the disparity in relaxation times is greater, with the 5 wt % gel exhibiting a  $\tau_{10}$  of 0.13 s compared with 0.45 s for the 10 wt % gel. Therefore, the gels of greater polymer concentration appear to have significantly slower relaxation times, except for voltages near the threshold. Since the polymer network dictates the long-time relaxation behavior, this indicates that the electric field modified the network morphology at voltages significantly higher than the threshold. Interestingly, the relaxation time is seen to decrease slightly with voltage at high voltages. This suggests the presence of competing interactions, which result in an increased relaxation time with voltage near the threshold but a decreased relaxation time at much higher voltages. At very large voltages, when the polymer network is significantly perturbed from equilibrium, it may have a larger restoring force once the applied voltage is removed.

Gels with a voltage previously applied tend to show faster switching and slower relaxation. At

Run	$ au_{90}~({ m s})$	$ au_{10}$ (s)
1	10.0547	0.0160
2	9.7084	0.0185
3	9.8387	0.0183
4	9.6162	0.0195
5	9.4906	0.0205
6	9.5682	0.0198
7	9.1146	0.0210
8	9.4956	0.0217
9	9.4956	0.0218
10	8.9699	0.0217

Table A.2: Results for successive transient voltage runs on 5 wt % LC gel in a 15  $\mu$ m gap. The voltage is at the threshold, 30 V.

voltages just above the threshold, there is very little effect on the subsequent electro-optic behavior of the gel (Tab. A.2). At twice the voltage, the  $\tau_{90}$  values decrease dramatically between the first and second experiments, between which the gel is allowed to relax for two minutes but is not reheated into the isotropic state (Tab. A.3). The response time steadily decreases in the subsequent experiments, but the rate of decrease is much smaller than that observed for the first and second experiments. This decrease in  $\tau_{90}$  with the application of a large voltage indicates that the modified morphology of the polymer network presents a decreased resistance to reorientation of the liquid crystal director along the electric field, and therefore the polymer network itself is being aligned with application of large voltages. At three times the voltage, both the  $\tau_{90}$  and the  $\tau_{10}$  values decrease further (Tab. A.4). The decrease in the  $\tau_{90}$  is more dramatic, by about an order of magnitude, and suggests that the applied field is dominating the reorientation time. The decrease in  $\tau_{90}$  may arise from a greater reorientation force when the polymer network is significantly perturbed from the morphology imprinted during crosslinking.

Run	$ au_{90}$ (s)	$ au_{10}$ (s)
1	3.7630	0.2508
2	0.1645	0.2388
3	0.0560	0.2715
4	0.0457	0.3116
5	0.0315	0.3621
6	0.0300	0.4120
7	0.0245	0.4630
8	0.0207	0.5201
9	0.0185	0.5581
10	0.0232	0.6009

Table A.3: Results for successive transient voltage runs on 5 wt % LC gel in a 15  $\mu m$  gap. The voltage is at twice the threshold, 60 V.

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Table A.4: Results for successive transient voltage runs on 5 wt % LC gel in a 15  $\mu m$  gap. The voltage is at three times the threshold, 90 V.

Run	$ au_{90}$ (s)	$ au_{10}$ (s)
1	0.1070	0.1342
2	0.0020	0.1213
3	0.0015	0.1418
4	0.0015	0.1620
5	0.0013	0.1841
6	0.0012	0.2040
7	0.0015	0.2298
8	0.0015	0.2505
9	0.0010	0.2723
10	0.0007	0.2936