A dual frequency addressed polymer stabilized pi-cell liquid crystal device

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Dual frequency addressing of nematic pi-cell devices produces submillisecond switching times since the liquid crystal can be driven both parallel and perpendicular to the applied field and there is no kick-back of the director during switching. The nucleation of the V state in devices containing dual frequency liquid crystal materials is much slower than that in conventional pi-cells, however. Polymer stabilization of the V state eliminates the need for nucleation each time the device is used. In this paper we present a polymer stabilized pi-cell containing a dual frequency liquid crystal material, and show that the presence of the polymer network significantly influences the switching of the device. Some optimization of the addressing scheme is required when switching the polymer stabilized device in order to avoid transient formation of the twisted state. Using this optimization, the switching time is under 3 ms across a wide range of addressing voltages.

1. Introduction

Dual frequency addressing of liquid crystal devices exploits the relaxation of the dielectric properties of nematic materials as the frequency of the applied field is increased. The relaxation of the permittivity along the long axis of the molecule ($\varepsilon_\parallel$) occurs at a much lower frequency than that of the permittivity perpendicular to the long axis ($\varepsilon_\perp$) [1]. If the dielectric anisotropy ($\Delta\varepsilon=\varepsilon_\parallel-\varepsilon_\perp$) is positive at d.c., this parameter can change sign as the frequency increases ($\varepsilon_\parallel$ relaxes while $\varepsilon_\perp$ remains constant). While all calamitic nematic liquid crystals experience this relaxation, in most it occurs at a few hundred kilohertz or more. *Dual frequency* liquid crystal materials are those in which the change in sign of $\Delta\varepsilon$ occurs at a few kilohertz. In such materials both the positive and negative dielectric anisotropy regimes are easily accessible.

The relaxation time of liquid crystal devices is much longer than the switching time during voltage switch-on. Dual frequency addressing allows the liquid crystal to be driven parallel to the applied field using a low frequency waveform in the positive $\Delta\varepsilon$ regime, and perpendicular to it using a high frequency waveform in the negative dielectric anisotropy regime. This technique has been used in an attempt to accelerate switching in a variety of device geometries [2–5]. Of particular interest is the dual frequency addressed pi-cell, since it has sub-millisecond switching times across a wide range of addressing voltages [5]. Such fast switching times are possible because there is no kick-back during switching from high tilt to low tilt states in pi-cells [6, 7], and this means that no rest period is required before application of the high frequency waveform (when switching devices in which kick-back effects are observed, rest periods are necessary to avoid re-orientation of the director in undesired directions [3, 8]).

2. The pi-cell

The pi-cell is a fast-switching nematic liquid crystal device [6] and is operated in the V state (see figure 1). Formation of the V state requires splayed surface pretilts and an applied voltage above the H to V state threshold, V (H–V). If there is no applied voltage, however, a splayed ground state forms (see figure 1). On sudden application of a voltage above the Fréedericksz tilt threshold, V (Fred), the symmetric H state forms [9], which then decays to one or other of the asymmetric H states over time [10, 11]. The two asymmetric H states are of equal energy, and so both will form in the device, with domain walls visible between them. If the voltage is applied gradually, then the symmetric H state is not seen, and the asymmetric H states form. These H states are not topologically similar to the V state, so when a voltage above V (H–V) is applied, nucleation is required to form the V state (the

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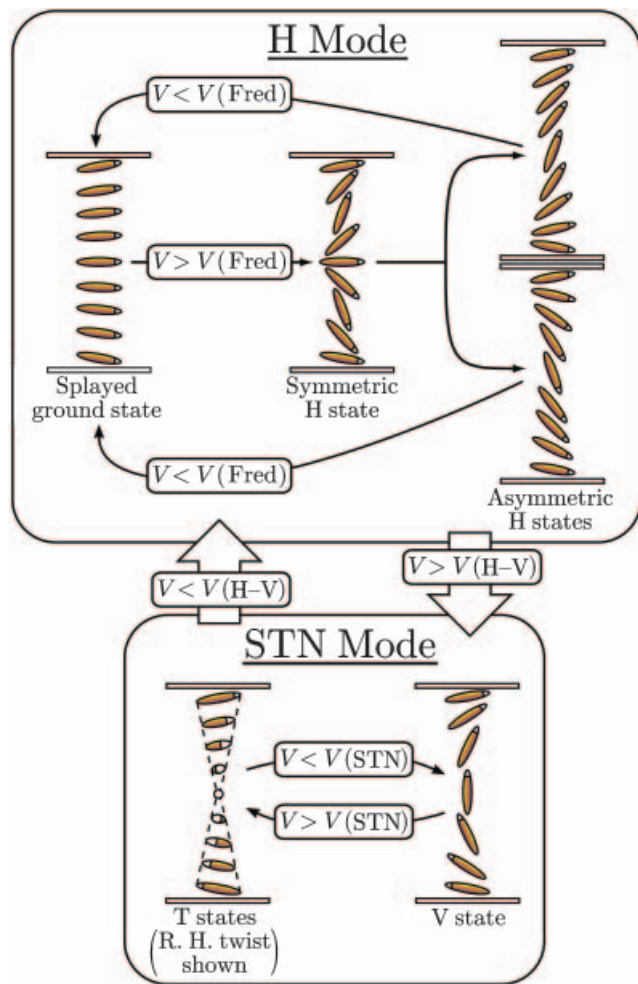
Figure 1.   The director configurations that form in splayed nematic devices as the applied voltage are varied. The symmetric H state only forms under sudden voltage application. The H and STN modes are not topologically similar, and so the transition from one to the other occurs through a nucleation process, indicated by dotted lines.

lower energy state must grow from a defect in the director structure). Once in the V state, if the voltage falls below the STN threshold, a 180° twisted state forms (the T state), and if the applied voltage is less than V (H–V), the H states will nucleate once again.

3.   V state nucleation in dual frequency pi-cells

Before the pi-cell can be operated, the V-state must be nucleated over the entire pixel area. In conventional pi-cells this is achieved by applying a voltage above V (H–V) in short pulses, and under these conditions the transition is from the symmetric H state to the V state [12]. Since the speed of domain growth is approximately proportional to the energy difference across the domain wall [13] and the symmetric H state is of much higher energy than the V state (see figure 2) the transition is
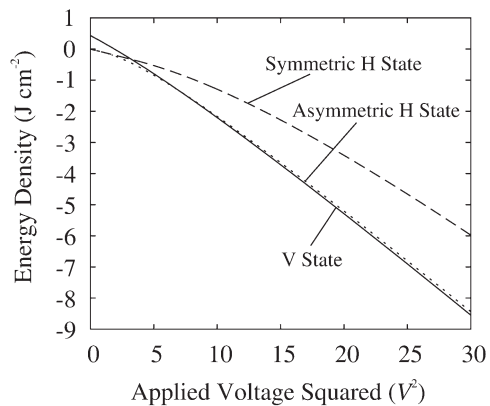

Figure 2.   Numerical simulation of the energies of the symmetric and asymmetric H states and the V state in a 2 μm thickness device filled with non-dual frequency, positive Δε material ZLI-1132 (see [11] for the material parameters and modelling technique). The large energy difference between the symmetric H state and the V state leads to very fast nucleation of the V state. When using dual frequency materials the symmetric H state does not form, and so the nucleation is from the asymmetric H states to the V state. This energy difference is much smaller, leading to a decrease in domain growth speed.

fast. If the applied voltage is not pulsed, the asymmetric H states form, and then the domain growth is far slower since the energy difference between these states and the V state is much lower (see figure 2).

When using dual frequency materials, however, the symmetric H-state does not form [5]. The domain growth is then between the asymmetric H states and the V state, regardless of whether the applied voltage is pulsed or not. The formation of the V state in dual frequency pi-cells is therefore much slower than that in conventional pi-cells. Figure 3 shows the nucleation of the V-state in a 4.4 μm thickness device filled with the dual frequency material TX-2A (Merck). A 1 kHz,

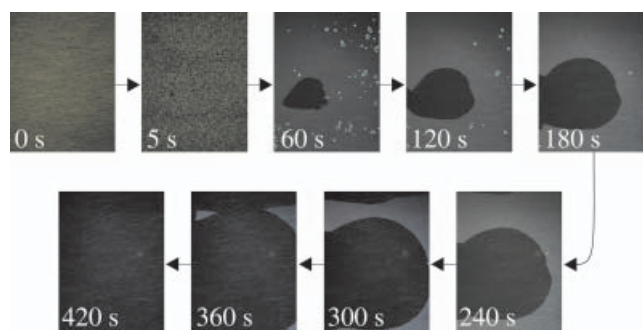Figure 3.   Nucleation of the V state in a 4.4 μm thickness pi-cell filled with the dual frequency mixture TX-2A (Merck) when a 1 kHz, 25 V_p sine wave is applied constantly. The nucleation process takes between six and seven minutes to complete.

$25\,V_p$ sine wave is applied suddenly at $t=0$, and many small asymmetric H state domains form, which appear bright in the photomicrographs. The V state takes several minutes to form in the entire pixel. Using non-dual frequency materials, this process would take only a few seconds using the pulsed voltage technique described above.

In order to produce relaxation of the dielectric properties at low frequencies, dual frequency materials must have a high rotational viscosity, and this will contribute to the slow nucleation of the V state when using this class of material. The relaxation time of a $4.5\,\mu m$ thickness pi-cell filled with TX-2A is around $8\,ms$ [5], while that of a pi-cell of the same thickness filled with non-dual frequency material ZLI-1132 is around $2\,ms$ [14], implying that the viscosity of the dual frequency material is approximately four times larger. As mentioned above, however, the transition to the V state when using dual frequency materials is far more than four times slower than when using non-dual frequency materials (a few minutes compared with a few seconds). By considering the relative energies of the director profiles, the transition from the symmetric H state to the V state is expected to be around ten times faster than that from the asymmetric H states to the V state [12]. This, combined with the increase in rotational viscosity, causes the slow nucleation of the V state when using dual frequency materials. Using the viscosity and energy difference adjustment factors quoted above, the V state transition time when using ZLI-1132 is predicted to be around $9\,s$, which is typical for a non-dual frequency material.

Figure 4 shows the domain growth of the V state around an asymmetric H-state domain wall. One asymmetric H state is present within the bright loop,
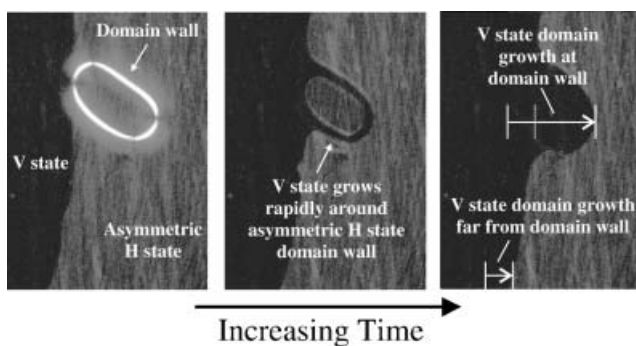

Figure 4.   V state domain growth around an asymmetric H state domain wall, which appears bright in the first photomicrograph. The V state domain growth is faster around the loop because the wall is of higher energy than the surrounding asymmetric H state. Note: the colour balance of the photomicrographs has been adjusted to clarify the distinction between the asymmetric H states and the V state.

and the other outside it (the darker region contains the V state). Since the asymmetric H state domain wall is of higher energy than the bulk, the V state grows very quickly around it. When a high voltage is applied suddenly to the device, many small asymmetric H state domains form, as can be seen in figure 3. The V state domain growth is fast under these conditions, since the asymmetric H state domain wall density is high. There is an energy cost associated with the domain walls, however, so isolated asymmetric H state domains shrink over time, leaving only a few large domains (in figure 3 most of the small domains have disappeared after $60\,s$). If the applied voltage is pulsed instead of applied constantly, however, the asymmetric H state domains remain small, leading to faster V state domain growth. This technique is demonstrated in figure 5, where a $1\,kHz$, $25\,V_p$ is applied in $8\,s$ pulses, with a $0.8\,s$ gap between pulses. The jagged appearance of the V-state domains after $60\,s$ in figure 5, compared with the smooth appearance shown in figure 3, clearly indicates that it is growth along the asymmetric H state domain walls similar to that shown in figure 4 that is accelerating the transition. The formation of the V state is faster under these conditions, with the whole pixel in this state after around $200\,s$.

4. Polymer stabilization of the V state

The length of time required to nucleate the V state in dual frequency pi-cells may be undesirable for some applications. In order to remove the need for nucleation every time the device is used, a polymer network can be used to stabilize the V state at zero applied voltage.

It has been shown that by adding small concentrations (2 to $10\,wt\%$) of reactive mesogens (RM) to conventional nematic liquid crystal materials, the V state can be stabilized [14, 15]. The RM is mixed with a
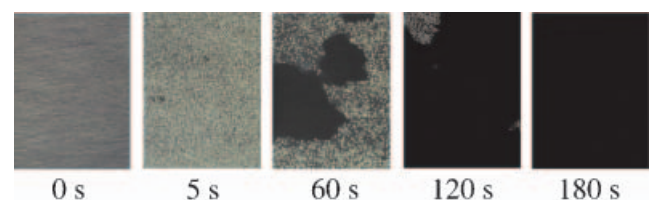

Figure 5.   Nucleation of the V state in a $4.4\,\mu m$ thickness pi-cell filled with the dual frequency mixture TX-2A when a $1\,kHz$, $25\,V_p$ sine wave is applied in $8\,s$ pulses with a gap of $0.8\,s$ between pulses. Pulsing of the applied waveform causes the asymmetric H state domains, which appear bright in the photomicrographs, to remain small. Since domain growth of the V state along the asymmetric H state domain walls is fast (see figure 4), this accelerates the formation of the V state. The V state is now present in the whole pixel after $200\,s$, decreasing the formation time by more than a factor of two.

photoinitiator, and then dissolved in the liquid crystal host. A device is filled with this mixture, and the V state is nucleated as described in §3. While applying a voltage just above V (H–V), the device is exposed to UV light, photopolymerizing the RM, and producing a sparse polymer network in the device. If a suffcient concentration of RM is used, the polymer network stabilizes the V state, even when the voltage is removed indefinitely (there is no H state nucleation and the twisted state does not form). This then eliminates the need to nucleate the V state each time the device is to be used — nucleation is required only when the device is manufactured.

5. Static response of the polymer stabilized pi-cell

A $4.4\,\mu$m pi-cell was filled with TX-2A containing $2\,$wt$\%$ of a 20:1 mixture of RM257 (Merck) and photoinitiator Irgacure 907 (Ciba). The RM was cured by exposing the device to UV light of wavelength $350\,$nm and intensity $\sim 30\,$W$\,$m^{-2} for $15\,$min while a $1\,$kHz, $4\,$V$_p$ sine wave was applied to retain the V state. Figure $6\,(a)$ shows the static response of the device to $1\,$kHz sine waves of different rms voltages (this is in the positive dielectric anisotropy regime). As the voltage is increased, the device switches to a higher tilt state reducing the effective birefringence of the device, and decreasing the transmission with the rubbing direction at $45°$ to crossed polarizers. Since the transmission is zero when the rubbing direction is parallel to one polarizer, the T state has not formed at any time (even with zero applied voltage). Thus, the polymer network has inhibited the static formation of the T state within the device.

The data in figure $6\,(b)$ show the static influence of applying a $50\,$kHz sine wave to the device (this is in the negative dielectric anisotropy regime). Clearly, the the T state forms as the voltage is increased since the transmission when the rubbing direction is parallel to one polarizer becomes non-zero.

6. Dynamic response of the polymer stabilized dual frequency pi-cell

The dynamic response of the polymer stabilized pi-cell to a dual frequency addressing scheme is shown in figure 7. For purposes of comparison, the elastic relaxation of the device is also shown. A *constant voltage* dual frequency addressing scheme is used: a $1\,$kHz sine wave at the addressing voltage is applied for $25\,$ms to switch to the high tilt state, and subsequently the frequency is changed to $50\,$kHz to switch back to the low tilt state. The length of the high frequency burst is adjusted to produce the fastest switching time possible.
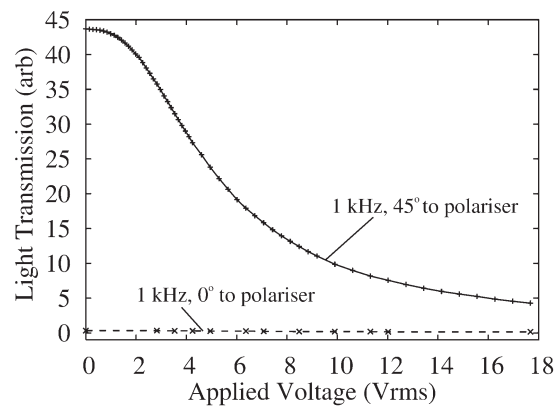
(a) Positive $\Delta\varepsilon$ regime
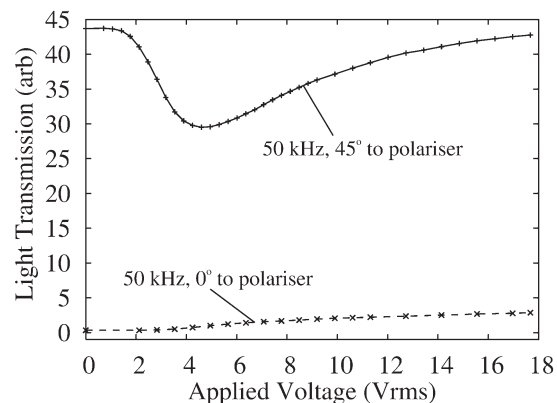
(b) Negative $\Delta\varepsilon$ regime

Figure 6. The static voltage–light transmission characteristics of the polymer stabilized dual frequency pi-cell under application of (a) $1\,$kHz and (b) $50\,$kHz sine waves. The results with the rubbing direction at $45°$ and parallel to one of the crossed polarizers are shown. If the transmission is non-zero with the rubbing direction parallel to the polarizers, then a twisted director profile is formed.

The schematic at the bottom of each plot in figure 7 indicates the timing of the pulses, with filled and unfilled boxes corresponding to the high and low frequency waveforms, respectively.

Figure $7\,(a)$ shows the device response when $5\,$V$_p$ waveforms are used, and the dual frequency addressing scheme significantly improves the switching time when compared with pure elastic relaxation of the same device. It is interesting to note that there is less oscillation after the application of the high frequency waveform than is observed in non-polymer stabilized dual frequency addressed pi-cells [5]. This indicates that the dynamic director profile produced by the high frequency waveform is more similar to the equilibrium director profile in the polymer stabilized device than in the non-polymer stabilized device. The switch-off time to the 90% switching level is $2.7\,$ms using the dual
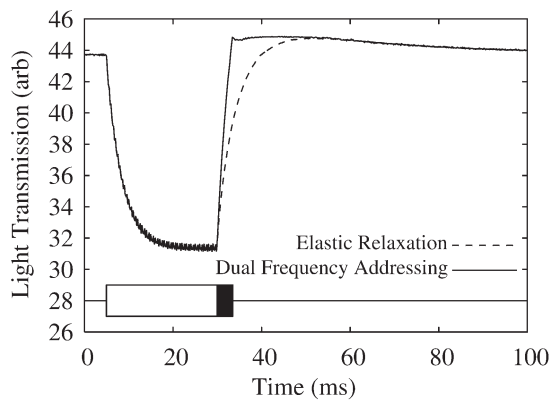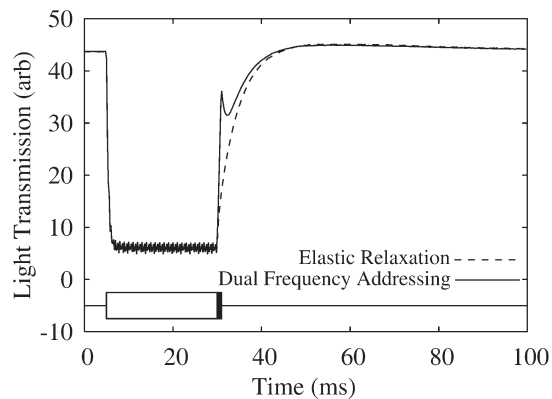
(a) 5 V$_p$ Waveforms

(b) 20 V$_p$ Waveforms

Figure 7. The dynamic response of the polymer stabilized dual frequency picell to (a) 5 V$_p$ and (b) 20 V$_p$ addressing schemes. A 1 kHz waveform is applied for 25 ms, after which the frequency is changed to 50 kHz while the voltage is kept constant. The length of the 50 kHz pulse is 3.5 ms for 5 V$_p$ and 1 ms for 20 V$_p$.

frequency addressing scheme, compared with 7.1 ms using elastic relaxation.

The device response to 20 V$_p$ amplitude addressing voltages is shown in figure 7 (b), and here the improvement in switching time with dual frequency addressing is much reduced. In this case the T state forms dynamically during the application of the high frequency pulse, producing the hump in transmission after the high frequency waveform has been removed. This T state formation means that a large amount of elastic relaxation is required to obtain the equilibrium director profile once the high frequency signal has been removed, increasing the switching time. The fact that the twisting occurs before the zero voltage transmission level has been reached indicates that the T state has formed before the tilt angle has reached its equilibrium level at zero applied voltage.
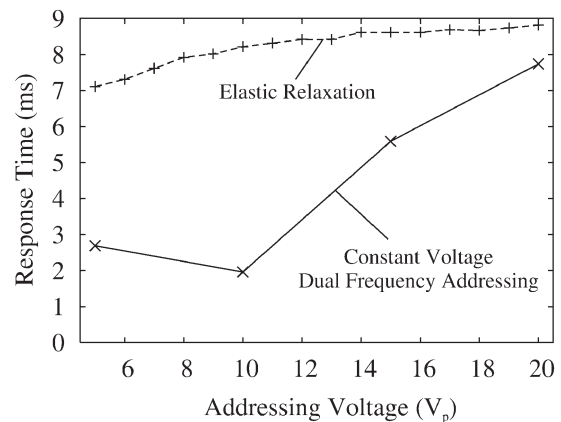

Figure 8. Switch-off times to the 90% switching level of the polymer stabilized device using the constant voltage addressing scheme (the voltages for the high and low frequency pulses are identical). The elastic relaxation time of the same device is included for comparison. With higher addressing voltages the dual frequency switching is inhibited by formation of the T state.

The switching times to the 90% level using the constant voltage addressing scheme and pure elastic relaxation are shown in figure 8. At lower voltages (10 V$_p$ and below) the dual frequency addressed switching times show significant improvement over the elastic relaxation time for the same device. As the voltage is increased, however, the dynamic formation of the T state reduces the benefit of using dual frequency addressing since the switching times are increased.

7. The influence of the polymer network on T state formation

The dynamic formation of the T state during dual frequency addressing is not observed in non-polymer stabilized dual frequency pi-cells [5]. Although the polymer network inhibits the static formation of the T state (there is no twist in the director profile with no applied voltage) it encourages dynamic formation of this state. This may be due to the inhomogeneity of the polymer network in the device; but this is unlikely, since the absorption of liquid crystal mixture TX-2A at the exposure wavelength (350 nm) is very small. The presence of the polymer network will still influence the visco-elastic properties of the device, however, altering the dynamic switching process significantly, causing the T state to form dynamically when using dual frequency addressing at high voltages.

8. Optimization of the polymer stabilized dual frequency pi-cell addressing scheme

The transient formation of the T state in polymer stabilized devices can be avoided by altering the

addressing scheme. Instead of using the constant voltage scheme described above, an amplitude of $10 V_p$ is always used for the high frequency burst, regardless of the voltage used for the 1 kHz addressing waveform. With a 50 kHz sine wave of this amplitude it takes more than 5 ms for the T state to form, and since the length of the high frequency burst required during device switch-off is less than this, formation of the T state can be avoided. Figure 9 shows the switching times to the 90% level using this optimized scheme, as well as the elastic relaxation times for the device. Clearly the new scheme has improved the switching times considerably, especially for the higher switch-on voltages.

9. Switching time comparison with non-polymer stabilized dual frequency pi-cells

It has been shown previously that polymer stabilized pi-cells are slower to switch than conventional pi-cells, and that this is probably due to inhibition of fluid flow by the polymer network [14]. A similar increase in switching time is seen in the polymer stabilized dual frequency device. Previously reported switching times for the non-polymer stabilized device with the constant voltage addressing scheme are around 1 ms for $10 V_p$ amplitudes [5]. From figure 8, the polymer stabilized device switches in around 2 ms under the same addressing conditions, which is commensurate with the increase in switching time caused by polymer stabilization in non-dual frequency pi-cells [14].

With no polymer network, high voltages can be used during switch-off to the low tilt state in dual frequency pi-cells, producing switching times of around 0.5 ms in a 4.4 μm thickness device containing TX-2A [5]. Such high voltages cannot be applied to the polymer stabilized device during switch-off, however, because of the transient formation of the T state during switching. This limit of the driving voltage, combined with the contribution from flow inhibition by the polymer network, increases the switching times of the polymer stabilized device to around 3 ms.

10. Conclusions

Dual frequency addressing of a polymer stabilized pi-cell has been demonstrated. Nucleation of the V state in devices filled with dual frequency liquid crystal materials is observed to be slow. The domain growth of the V state can be accelerated, however, by pulsing the applied voltage which produces very small asymmetric H state domains. Since the V state domain growth rate along the domain walls between the two asymmetric H states is fast, this decreases the transition time to the V state significantly. V state nucleation in a 4.4 μm thickness device still takes several minutes to complete when using this technique, however. The nucleation process can be avoided by stabilizing the V state using a sparse polymer network. This network, however, causes the 180° twisted state to form during switching when large amplitude high frequency waveforms are used. Optimization of the addressing scheme by limiting the amplitude of the high frequency waveform produces switching times below 3 ms across the whole range of addressing voltages. Since the device can be driven to both lower and higher tilted states with the applied field, the switching time between arbitrary grey scales should be of a similar magnitude.
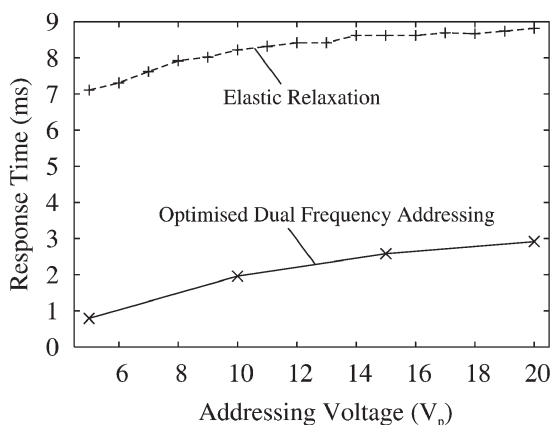
Acknowledgements

Figure 9. Switch-off times to the 90% switching level for the polymer stabilized device using the optimized addressing scheme (the voltage of the 50 kHz waveform is always $10 V_p$). The elastic relaxation time of the same device is included for comparison.

References

[1] M. Schadt. *Mol. Cryst. liq. Cryst.*, **89**, 77 (1982).
[2] E.P. Raynes, I.A. Shanks. *Electron. Lett.*, **10**, 114 (1974).
[3] I.A. Shanks, M.G. Clark, F.M. Leslie. U.S. Patent 4 328 493 (1984).
[4] Y.Q. Lu, X. Liang, Y.H. Wu, F. Du, S.T. Wu. *Appl. Phys. Lett.*, **85**, 3354 (2004).
[5] P.D. Brimicombe, S.J. Elston, E.P. Raynes. *Appl. Phys. Lett.* (submitted).
[6] P.J. Bos, K.R. Koehler/Beran. *Mol. Cryst. liq. Cryst.*, **113**, 329 (1984).
[7] H.G. Walton, M.J. Towler. *Liq. Cryst.*, **27**, 1329 (2000).

[8] P.D. Brimicombe, L.A. Parry-Jones, S.J. Elston, E.P. Raynes. *J. appl. Phys.*, **98**, 104104 (2005).

[9] M.J. Towler, E.P. Raynes. In Proceedings of the 22nd International Display Research Conference (Eurodisplay '02), Nice, France, Vol. 2, 877–881 (2002).

[10] L.Z. Ruan, J.R. Sambles. *Appl. Phys. Lett.*, **86**, 052502 (2005).

[11] P.D. Brimicombe, E.P. Raynes. *Liq. Cryst.*, **32**, 1273 (2005).

[12] H. Nakamura, M. Noguchi. *Jpn. J. appl. Phys.*, **39**, 6368 (2000).

[13] E.J. Acosta, M.J. Towler, H.G. Walton. *Liq. Cryst.*, **27**, 977 (2000).

[14] P.D. Brimicombe, S.J. Elston, E.P. Raynes. *Mol. Cryst. Liq. Cryst.* (submitted).

[15] S.H. Kim, L.C. Chien. *Jpn. J. appl. Phys.*, **43**, 7643 (2004).