# First-Order Fréedericksz transition in a Liquid-Crystal-Light-Valve.

M.G. Clerc*, T. Nagaya†, S. Residori** and C. S. Riera‡

*Departamento de Física, Facultad de Ciencias Físicas y Matemáticas, Universidad de Chile, casilla 487-3, Santiago, Chile
†Faculty of Education, Okayama University, Okayama, 700-8530, Japan
**Institut Non Linéaire de Nice, UMR 6618 CNRS-UNSA, 1361Route des lucioles, F-06560 Valbonne-Sophia Antipolis, France
‡Department of Mechanical Engineering, Massachussetts Institute of Technology,77 Massachussetts Avenue, Cambridge MA 02139, USA

**Abstract.** Light feedback in a Liquid-Crystal-light-Valve can render the Fréedericksz transition a first order one. A theoretical and experimental study of this first order transition is presented. An amplitude equation is derived, valid close to the transition. Depending on the values of the feedback parameters, both theory and experiment exhibit bistability, propagation of fronts and a Maxwell point.

## INTRODUCTION

One of the most well-studied phenomena in the physics of liquid crystals is the field-induced distortion of a nematic liquid-crystal film, called the Fréedericksz transition (FT) [1]. For a nematic film, FT is usually a second-order transition [2]. The possibility of modifying FT into a first-order one has been considered, either through the simultaneous application of electric and magnetic field [3] or through the action of an optical field [4], for liquid crystals possessing a very large optical anisotropy. In both cases, an experimental verification is difficult to attain, especially over a large area of the liquid crystal film. Another approach, is to realize a global feedback by means of spatially integrated light intensity [5]. In this case, the FT experimentally displays clear features of a first-order transition. However, spatial dynamics are lost as a consequence of integration.

By means of a a Liquid Crystal Light Valve (LCLV), we have realized an experiment in which the local feedback is high enough to render FT first-order over a large size ($\simeq cm^2$) of the nematic film [6]. Thus, all the spatial phenomena that we expect to be associated with a first-order phase transition can be observed and investigated. In particular, the classical nucleation theory applies, and we show that a local perturbation leads to a front propagation which, depending on the distance from the Maxwell point, is associated to shrinking or expanding droplets.

*Experimental setup.* A LCLV consists essentially in a nematic liquid-crystal film sandwiched between a glass and a photoconductive plate over which a dielectric mirror is deposed. Coating of the two bounding plates induces a planar anchoring (nematic director $\vec{n}$ parallel to the walls) of the liquid crystal film. Moreover, transparent electrodes

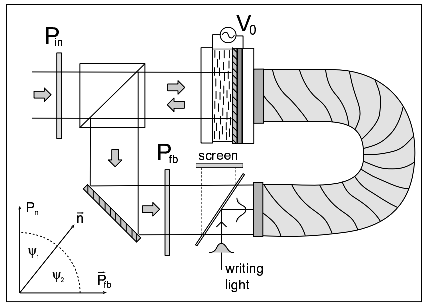


**FIGURE 1.** Experimental setup. Two confocal lenses, (not displayed in the scheme), provide a 1 : 1 image-forming system from the front side of the LCLV to its rear side. The optical feedback loop is closed by a fiber bundle, which is aligned in order to avoid any rotation or shift. $P_{in}$ and $P_{fb}$ are, respectively, the input and feedback polarizer. Their orientation with respect to the liquid crystal director is indicated in the left bottom of the figure.

covering the two plates permit the application of an electric field across the liquid-crystal layer. The photoconductor behaves like a variable resistor, which resistance decreases for increasing illumination. The light-driven feedback is obtained by sending back onto the photoconductor the light which has passed through the liquid-crystal layer and has been reflected by the dielectric mirror. The voltage $V_0$ applied to the LCLV is sinusoidal and of frequency $f = 20\ KHz$, which is much larger than the liquid crystal response time.

The experimental setup, displayed in Fig.1, consists of a LCLV with optical feedback, [7]. Here, the optical loop is designed in such a way that the light beam experiences no diffraction as well as no geometrical transformation, so that no transverse structures are formed close to the FT. The light intensity $I_w$ reaching the photoconductor is given by [8]

$$I_w = |\cos(\psi_1)\cos\psi_2 + \sin\psi_1 \sin\psi_2 e^{-i\beta cos^2\theta})|^2 I_{in} \tag{1}$$

where $\theta$ is the liquid crystal re-orientation angle, $\beta \equiv 2kd\Delta n cos^2\theta$ is the overall phase shift experienced by the light traveling forth and back through the liquid crystal layer, $I_{in}$ is the input intensity, $\psi_1$ and $\psi_2$ are the angles formed by the input and feedback light polarization with the liquid crystal director $\vec{n}$, respectively, $k = 2\pi/\lambda$ is the optical wavenumber, $d$ is the thickness of the liquid crystal layer and $\Delta n$ is the difference between the extraordinary ($\parallel$ to $\vec{n}$) and ordinary ($\perp$ to $\vec{n}$) index of refraction of the liquid crystal. In our experiment, $\beta \simeq 120$ since $\lambda = 633\ nm$, $\Delta n = 0.2$, $d = 30\ \mu m$. Input light intensity is typically around $I_{in} \simeq 0.9\ mW/cm^2$.

## THEORETICAL DESCRIPTION

*Derivation of the normal form.* For a nematic liquid crystal layer on which an electric field is applied, the dynamical equation for the director reads [2]

$$\gamma\vec{n} \wedge \partial_t \vec{n} = -\vec{n} \wedge \frac{\delta\mathcal{F}}{\delta\vec{n}}, \quad \vec{n}.\vec{n} = 1 \tag{2}$$

where $\gamma$ is the rotational viscosity, and $\mathcal{F}$ the Frank free energy which is expressed by

$$\begin{aligned} \mathcal{F} &= \frac{1}{2}\int K_1(\vec{\nabla}.\vec{n})^2 + K_2(\vec{n}.(\vec{\nabla}\wedge\vec{n}))^2 + K_3(\vec{n}.(\vec{\nabla}\wedge\vec{n}))^2 \\ &\quad -\varepsilon_\perp \vec{E}^2(\vec{n}) - \varepsilon_a(\vec{n}.\vec{E}(\vec{n}))^2 d\vec{r}. \end{aligned} \tag{3}$$

$K_i$ are the elastic constants of the liquid crystal, $\varepsilon_a$ the dielectric anisotropy and $\varepsilon_\perp$ the perpendicular dielectric permeability. For the sake of simplicity, we assume $K_1 = K_2 = K_3 = K$, leading to

$$\begin{aligned} \gamma\partial_t\vec{n} &= K(\nabla^2\vec{n} - \vec{n}(\vec{n}.\nabla^2\vec{n})) + \varepsilon_a(\vec{n}.\vec{E})(\vec{E} - \vec{n}(\vec{n}.\vec{E})) + \tfrac{\varepsilon_\perp}{2}\tfrac{\partial\vec{E}^2}{\partial\vec{n}} \\ &\quad -\tfrac{\varepsilon_\perp}{2}(\vec{n}.\tfrac{\partial}{\partial\vec{n}})\vec{E}^2 + \tfrac{\varepsilon_a}{2}(\vec{n}.\vec{E})\vec{n}.((\vec{n}.\tfrac{\partial}{\partial\vec{n}})\vec{E}). \end{aligned} \tag{4}$$

The total electric field applied to the liquid crystal layer depends on the response of the photoconductor to the write intensity $I_w$ and to the voltage applied to the liquid crystal layer ($V_0 = E_0/d$). As the light intensity is sufficiently small, the response of the photoconductor is linear. Thus, the total electric field reads $E(n) = E_0 + \alpha I_w(\vec{n})$, where $\alpha \simeq 4$ is a phenomenological parameter that we can evaluate from the experimental characteristics of the LCLV, (measuring $I_w$ in $mW/cm^2$).

After substituting (**??**) in the expression for the electric field, it becomes $E(n) = E_0 + A + B\cos(\beta\cos^2\theta)$, where $A = \frac{1}{4}(\cos 2(\psi_1 - \psi_2) + \cos 2(\psi_1 + \psi_2) + 2)\alpha I_{in}$ and $B = \frac{1}{4}(\cos 2(\psi_1 - \psi_2) - \cos 2(\psi_1 + \psi_2))\alpha I_{in}$. Since the liquid crystals are aligned parallel to the walls (planar anchoring) and are submitted to a perpendicular electric field, the electric field is $\vec{E} = (0,0,E)$ and $\vec{n} = (n_x, 0, n_z)$ with $n_x^2 + n_z^2 = 1$. By inserting $E(n)$ in (**??**) and by means of standard bifurcation theory, we derive an amplitude equation for the first unstable Fourier mode, $n_z = u(x,y)\sin(\pi z/d)$ and $n_x = 1 - u^2\sin^2(\pi z/d)/2$, which describes the director orientation at the onset of the instability. The equation reads

$$\partial_t u = c_1 u + c_3 u^3 + c_5 u^5 + K\nabla_\perp^2 u \tag{5}$$

where the coefficients $c_1$, $c_3$ and $c_5$ are given by

$$\begin{aligned} \gamma c_1 &= -\tfrac{K\pi^2}{d^2} + (A + E_0 + B\cos(\beta))(\varepsilon_a(A + E_0) + B(\varepsilon_a\cos(\beta) + 2\varepsilon_\perp\beta\sin(\beta))) \\ \gamma c_3 &= \tfrac{1}{3}\tfrac{K\pi^2}{d^2} - \tfrac{2}{3}\varepsilon_a(E_0 + A)^2 + \tfrac{2}{\pi^2}\varepsilon_\perp(2B\beta\sin(\beta))^2 - \tfrac{2}{3\pi^2}(12\beta^2\varepsilon_\perp + \pi^2\varepsilon_a)(B\cos(\beta))^2 \\ &\quad + \tfrac{\beta}{\pi^2}(\varepsilon_a(8+\pi^2) - \varepsilon_\perp\pi^2)(A + E_0 + B\cos(\beta))B\sin(\beta) \\ &\quad - \tfrac{4}{3\pi^2}(6\beta^2\varepsilon\perp + \pi^2\varepsilon_a)(E_0 + A)B\cos(\beta)) \\ \gamma c_5 &= (\tfrac{2B\beta}{\pi^2})^2((4+\pi^2)\varepsilon_a - \pi^2\varepsilon_\perp)(\sin(\beta)^2 - \cos(\beta)^2) \\ &\quad - \tfrac{B\beta}{12\pi^4}(192\beta^2\varepsilon_\perp + (9\pi^2 + 64)\pi^2\varepsilon_a)(E_0 + A)\sin(\beta) - (\tfrac{2\beta}{\pi^2})^2((4+\pi^2)\varepsilon_a - \pi^2\varepsilon_\perp)(E_0 \\ &\quad + A)B\cos(\beta) - \tfrac{B^2}{12\pi^4}(9\pi^2 + 64)\varepsilon_a + 768\beta^2\varepsilon_\perp)\beta\sin(\beta)\cos(\beta). \end{aligned} \tag{6}$$

*Discussion.* When $c_3$ is negative and of order one, Eq. (**??**) describes a second order FT. The FT becomes first order when $c_1$ and $c_3$ are positive (and small), with $c_5$ negative.

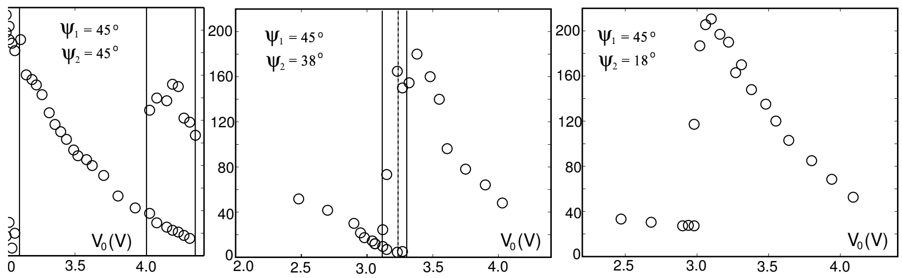


**FIGURE 2.** $I_w$ as a function of the r.m.s. values of the applied voltage $V_0$, for different angles, $\psi_1$ and $\psi_2$, between the polarizers and the liquid crystal director.

Qualitatively, this situation well describes the experimental observations. $c_3$ is a function of the polarizer angles, of the input intensity and of the voltage applied to the LCLV. In particular, we have verified that changes in the polarizer angles produce effects with the same sign in the theory and in the experiment. However, it is important to underline that the theoretical description of the transition is valid only in the asymptotic limit $c_3 \sim (c_1)^{1/2}$. Instead, a quantitative description of the experiment would require $c_3 \sim 1$, which cannot be obtained from an asymptotic analysis [9]. Henceforth we consider $c_3 > 0$ and $c_5 < 0$.

## RESULTS AND COMPARISON WITH THE MODEL

In Fig.2 we show $I_w$ as a function of $V_0$ and in the presence of feedback. Three bifurcation diagrams are reported, for three different values of the polarizer angles $\psi_1$ and $\psi_2$. For $\psi_1 = \psi_2 = 45^0$, which maximizes the birefringence of the LCLV, the diagram displays an abrupt change at the transition point and an hysteresis region. For this value of $\psi_1$ and $\psi_2$, the extension of the hysteresis region is maximum, in agreement with the model. Notice that $I_w$ is measured by a small area photodiode, i.e., it is a local measurement taken at the center of the feedback beam. By looking at the entire image of the feedback beam with a CCD camera, we see that the transition point is characterized by a white spot developing over a dark background. In the course of time, the front of the white spot expands and the white state overcomes the dark one. For the amplitude equation, this represents a front solution between the unstable state $u_0 = 0$ and the stable state $u_{\pm}^2 = (-c_3 \pm \sqrt{c_3^2 - 4c_1c_5})/(2c_5)$, a Kolmogorov-type front [10]. By further increasing the voltage, the LCLV birefringence changes and the white state becomes ”grey” until the dark value is reached again. Successive transitions to the white state are present for larger values of the applied voltage. These states correspond to parameter values far from the FT, and hence they are not considered here.

By decreasing the voltage, we observe a hysteresis cycle. In order to determine the size of the bistable region, we inject an additional light spot (low power He-Ne laser) into the feedback loop. This acts as a small perturbation, triggering the transition from

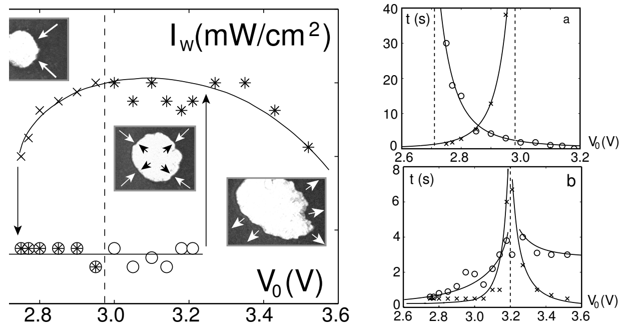


**FIGURE 3.** Bistability and front propagation. On the left, $I_w$ *vs* $V_0$ plot; circles: dark state with writing light off; stars: white state with writing light off; cross: white state with writing light on. On the right, a) $\tau_r$ (circles) and $\tau_f$ (cross); b) $\tau_{on}$ (cross) and $\tau_{off}$ (circles) as a function of $V_0$.

the dark state to the white one. The white state persists when we block the additional writing light, while it switches to the dark state if we perturb the feedback. In Fig.2, the arrows delimit the region over which this writing-erasing procedure is robust. Notice that spatial inhomogeneities and other noise sources can influence the stability of the two states. Furthermore, just before the transition, there is an appreciable decrease of the intensity $I_w$. This is indeed a signature of the fact that we are approaching the transition point, where fluctuations become very large and light is diffused in all directions, in the same way as it occurs at the critical point for a liquid-vapor transition. This sort of critical opalescence [11], is responsible for the diminished efficiency of the light reflection from the LCLV.

For $\psi_1 = \psi_2 = 45^0$, a more detailed set of measurements is reported in Fig.3, left side, for a $V_0$ range centered around the transition point and together with three representative images of the feedback field. These images show the direction of the front propagation, depending on the mutual stability of the white and the dark state. The dashed line marks the point at which the front propagation is zero, usually called the Maxwell point. Below this point the white state is less stable than the dark one and the white spot, once created by the writing light, contracts to zero. Above the FT point, the white spot nucleates spontaneously and its front expands until the white state covers all the background. In between, the front of the spot expands or retracts depending on the size of the perturbation.

The three crucial points, i.e., the beginning of the bistability, the Maxwell point and the FT point, are identified by the divergence of the response times. We show in Fig.3, right side, the response times associated with the bifurcation diagram. In a, we show the rise time $\tau_r$ needed for a white spot to develop once the writing light is switched on and the fall time $\tau_f$ taken by the white spot to disappear when the writing light is switched off. The divergence of the first one identifies the beginning of the bistable region whereas the divergence of the second one corresponds to the Maxwell point. In b, we show the rise $\tau_{on}$ and fall $\tau_{off}$ time when the feedback is switched respectively on and off. The divergence of these two times corresponds to the FT point. Correspondingly, in the amplitude equation (**??**), the beginning of the bistability is characterized by $c_3^2 - 4c_1c_5 \geq 0$, $c_1 < 0$, the Maxwell point by $c_3^2 = 16c_5c_1/3$ $(c_1 < 0)$ and the FT by

$c_1 = 0$.

## CONCLUSION

In conclusion, we have derived an amplitude equation valid close to the FT for a nematic liquid crystal layer in the presence of a light-driven feedback. We have shown that, as a consequence of feedback, FT becomes first order. Our theoretical description is in a fair qualitative agreement with the experimental observations. The feedback is provided by the conversion of optical intensity into electric field via a photoconductive transducer (LCLV). Bistability is very robust here and is present for a relatively wide range of the experimental parameters. Spatial dynamics of front propagation is also reported.

## ACKNOWLEDGMENTS

This work has been supported by the ACI Blanche of the French Ministry of Research. M. Clerc has benefited from the financial support from the Centro de Fisica No Lineal y Sistemas de Santiago. C. Riera was supported as a postdoctoral fellow by an ONR- NYI grant awarded to L. Mahadevan at the Massachusetts Institute of Technology (2000-03).

## REFERENCES

1. V. Fréedericksz and V. Zolina, Trans. Faraday Soc. **29**, 919 (1933).
2. P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, (Oxford Science Publications, Clarendon Press, second edition, 1993).
3. B.J. Frisken and P. Palffy-Muhoray, Phys. Rev. A **39**, 1513 (1989), Phys. Rev. A. **40**, 6099 (1989); S. Garg, S. Saeed and U.D. Kini, Phys. Rev. E **51**, 5846 (1995).
4. S.D. Durbin, S. M. Arakelian and Y.R. Shen, Phys. Rev. Lett. **47**, 1411 (1981); A. J. Karn, S.M. Arakelian, Y.R. Shen and H. L. Ong, Phys. Rev. Lett. **57**, 448 (1986).
5. Peng-Ye Wang, Hong-ju Zhang and Jian-hua Dai, Opt. Lett. **12**, 654 (1987).
6. M.G. Clerc, S. Residori and C.S. Riera, Phys. Rev. E **63**, 060701(R) (2001).
7. S.A. AKhmanov, M.A. Vorontsov and V.Yu. Ivanov, JETP Lett. **47**, 707 (1988).
8. see e.g. F.T. Arecchi, S. Boccaletti and P.L. Ramazza, Physics Reports **318**, 1 (1999) and references therein.
9. P. Manneville, *Dissipative structures and weak turbulence*, (Academic Press, San Diego 1990).
10. M. Cross and P. Hohenberg, Rev. Modern Phys. **65**, 581 (1993).
11. L. Mandel and E. Wolf, *Optical Coherence and quantum Optics*,(Cambridge University Press, New York, 1995).