



## Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and  
subscription information:

<http://www.tandfonline.com/loi/gmcl18>

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Version of record first published: 24 Sep 2006.

To cite this article: Ru-Pin Pan, Shyu-Mou Chen & Ting-Chiang Hsieh (1991): The Response Time and the Backflow Effect of 5CB and 8CB Films with a Free Surface in Magnetic Fields, *Molecular Crystals and Liquid Crystals*, 198:1, 99-106

To link to this article: <http://dx.doi.org/10.1080/00268949108033387>

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# The Response Time and the Backflow Effect of 5CB and 8CB Films with a Free Surface in Magnetic Fields

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*(Received July 25, 1990)*

The dynamic response of 5CB and 8CB films with a free surface in an external magnetic field was investigated. The magnetic field was initially above the threshold for the Freedericksz transition. Two kinds of time constants were measured optically: one is the turn off time constant when the magnetic field was reduced to a value below the threshold, and the other is the response time with the magnetic field slightly changed but still above the threshold.

The results for samples with various thickness were compared with those films sandwiched between two glass substrates. The differences were analyzed with backflow taken into account. A satisfactory fit with the theory was obtained. Several viscosity coefficients for these two liquid crystal materials have also been determined.

*Keywords: response time, backflow effect, free surface, viscosity*

Surface effects of liquid crystals are interesting for both areas of device applications and basic understanding of physical phenomena. The study of free surface effects is even more interesting for understanding the anisotropy of molecular interactions. Previous studies have shown the existence of preferred molecular orientation at the free surface of nematic liquid crystals (NLC).<sup>1,2</sup> Ru-pin Pan *et al.*,<sup>3</sup> have reported the Freedericksz transition<sup>4</sup> and relaxation response of 4'-*n*-pentyl-4-cyanobiphenyl (5CB) nematic liquid crystal films with a free surface induced by a laser field. In this work, a dc magnetic field is used to induce molecular reorientation to avoid the thermal and finite beam size effects caused by a laser beam. It has been shown<sup>5</sup> that the critical transition magnetic field  $H_c$  of NLC of 5CB and 8CB (4'-*n*-octyl-4-cyanobiphenyl) films with a free surface (FS) under the magnetic field is the same as for the similar hard bounded films sandwiched between glasses (HB). This observation has been interpreted as a strong surface anchoring at the free surface, the boundary condition for reorientation is therefore similar to the hard boundary, that is  $\Delta\theta = 0$ .

Although the static angular distortion is same for the FS and HB films with same thickness due to the same boundary conditions and same free energy equation, the molecular flow can be much different at the free surface due to the lacking of positional anchoring. Since the orientational motion is coupled with flow motion,

the boundary condition on flow can alter the rotational speed, thus the response time will be different for FS and HB films. Here, we present our studies on the response time of both HB and FS samples when the external magnetic field  $\vec{H}$  is turned off or slightly varied.

If the external magnetic field  $H$  is changed abruptly from an initial field  $H_1$  smaller than the critical field,  $H_c$ , to a field  $H$  larger than  $H_c$ , the liquid crystal molecules will begin to rotate, we define the dynamic exponential time constant at the beginning as the turn-on time constant,  $\tau_{on}$ . Similarly, if the magnetic field is changed from  $H_2 > H_c$  to  $H < H_c$ , the molecular orientation will relax to  $\theta = 0$ . We define the exponential time constant at the end as the turn-off constant,  $\tau_{off}$ . Following the work by Pieranski *et al.*,<sup>8</sup> we take  $\tau_{on}$  to be positive and  $\tau_{off}$  to be negative.

For small orientation angle  $\theta$ , the coupled equations of motion for the director  $\hat{n} = (\theta, 0, 1)$  and the flow velocity  $\vec{v} = (v_x(z), 0, 0)$  can be written as<sup>8</sup>:

$$k_{33} \frac{\partial^2 \theta}{\partial z^2} + \chi_a H^2 \theta = \gamma_1 \frac{\partial \theta}{\partial t} + \alpha_2 \frac{\partial v_x}{\partial z}, \quad \text{and} \quad \frac{\partial}{\partial z} \left( \eta_c \frac{\partial v_x}{\partial z} + \alpha_2 \frac{\partial \theta}{\partial t} \right) = 0. \quad (1)$$

where  $\eta_c \equiv 1/2 (\alpha_4 + \alpha_5 - \alpha_2)$ ,  $\gamma_1 \equiv \alpha_3 - \alpha_2$  and  $\alpha_1, \dots, \alpha_6$  are the viscosity coefficients following the notations of Reference (4).

The time constant for HB film,  $\tau_{hb}(H)$ , is obtained by solving Equation (1) with a trial solution satisfying the boundary conditions,  $\theta = v_x = 0$ , at  $z = \pm d/2$ ,<sup>6</sup>

$$\tau_{hb}^{-1}(H) = \chi_a (H^2 - H_c^2) / \gamma_{1,hb}^*(h), \quad (2a)$$

where  $\gamma_{1,hb}^*(h)$  is an effective viscosity and is a function of  $h = H/H_c$ . For small fields, the relative variation of  $\gamma_{1,hb}^*$  with  $h$  is negligible, and

$$\gamma_{1,hb}^* \equiv \left( 1 - \frac{1}{6} \alpha_2^2 / \gamma_1 \eta_c \right) \gamma_1 < \gamma_1. \quad (2b)$$

At the free surface, the boundary conditions for FS samples are:

$$\theta = v_x = 0, \text{ at } z = -d/2; \quad \text{and} \quad \theta = \frac{\partial v_x}{\partial z} = 0, \text{ at } z = +d/2. \quad (3)$$

With a proper trial solution,<sup>5</sup> which satisfies the boundary conditions, a similar equation for the response time constant of FS samples,  $\tau = \tau_{fs}(H)$ , is obtained,

$$\tau_{fs}^{-1}(H) = \chi_a (H^2 - H_c^2) / \gamma_{1,fs}^*, \quad \text{where} \quad \gamma_{1,fs}^* = (1 - \alpha_2^2 / \gamma_1 \eta_c) \gamma_1. \quad (4)$$

If the applied magnetic field is turned off from an initial field larger than  $H_c$ , the zero field turn off rate  $\tau^{-1}(0)$  is then

$$\tau^{-1}(0) = -(k_{33} / \gamma_1^*) (\pi^2 / d^2), \quad (5)$$

where  $\gamma_1^* = \gamma_{1,hb}^*$  or  $\gamma_{1,fs}^*$  for HB or FS samples, respectively.

We define the relaxation time constant for small variation in angle,  $\Delta\theta$ , around the equilibrium angle,  $\theta$ , in a magnetic field  $H$  as the differential time constant. In general,  $\theta$  is not a small value. The simplified equations of motion for small angles are no longer valid. We must start from more general equations of motion to derive the differential time constant,  $\tau_d$ . The result for differential time constant is summarized as follows.

For HB films,

$$\tau_{d,hb} = \left(1 + \frac{B}{A_{hb}} \frac{\gamma_{hb}}{\gamma_1}\right) \tau_{do}, \quad (6)$$

and for FS samples,

$$\tau_{d,fs} = \left(1 + \frac{B}{A_{fs}} \frac{\gamma_{fs}}{\gamma_1}\right) \tau_{do}, \quad (7)$$

where  $\tau_{do}$  is the differential time constant without considering the backflow effect, and

$$\tau_{do} = \gamma_1 \{q^2 [K_{33} + (K_{11} - K_{33})G_1] + \chi_a H^2 G_2\}^{-1}$$

$$\gamma_{hb} = \frac{1}{2} (\gamma_1 - \gamma_2)(1 - 8/\pi^2) + \gamma_2 G_3, \quad \gamma_{fs} = \frac{1}{2} (\gamma_1 + \gamma_2 G_2),$$

$$A_{hb} = \eta_c + \alpha_1 G_4 + \gamma_2 G_5 + \alpha_1 G_6 + \gamma_2 G_7, \quad A_{fs} = \eta_c + \alpha_1 G_4 + \gamma_2 G_5,$$

$$B = \alpha_2 - \gamma_2 G_5,$$

$$G_1 = \sum_{n=0}^{\infty} (-1)^n \frac{2n+3}{4(n+1)(2n+1)!} (2\theta_m)^{2(n+1)} p_n,$$

$$G_2 = \sum_{n=0}^{\infty} (-1)^{n+1} \frac{2}{(2n)!} (2\theta_m)^{2n} p_n,$$

$$G_3 = \sum_{n=0}^{\infty} (-1)^n \frac{1}{2(n+1)!} (2\theta_m)^{2(n+1)} \left( p_{n+1} - \frac{2}{\pi^2(n+2)p_n} \right),$$

$$G_4 = \sum_{n=0}^{\infty} (-1)^n \frac{2n+3}{8(2n+2)!} (4\theta_m)^{2(n+1)} p_n,$$

$$G_5 = \sum_{n=0}^{\infty} (-1)^n \frac{2n+3}{2(2n+2)!} (2\theta_m)^{2(n+1)} p_n,$$

$$G_6 = \frac{1}{2\pi^2} \sum_{n=0}^{\infty} (1-1)^{n+1} \frac{1}{(2n+2)!} (4\theta_m)^{2(n+1)} p_n^{-1},$$

$$G_7 = \frac{2}{\pi^2} \sum_{n=0}^{\infty} (-1)^{n+1} \frac{1}{(2n+1)!} (2\theta_m)^{2(n+1)} p_n^{-1},$$

and

$$p_n = \frac{1}{2} \frac{3}{4} \frac{5}{6} - \frac{2n+1}{2n+2}.$$

The derivation can be found in Reference 5. Equations (6) and (7) are consistent with  $\tau_{\text{off}}$  at magnetic field  $H$  smaller than the critical field  $H_c$ .

The material 5CB & 8CB used in our experiments were purchased from British Drug Houses, Inc. Their nematic phase ranges are 22.5 to 35.2°C and 33.5 to 40.2°C (Reference 7), respectively. The preparation procedure of the samples have been reported elsewhere.<sup>3</sup>

For both HB and FS samples, the film thickness range from 125  $\mu\text{m}$  to 300  $\mu\text{m}$ . These are measured either mechanically with a probe pin<sup>3</sup> or interferometrically within an accuracy about  $\pm 2 \mu\text{m}$ . Detailed description of these methods has been presented in a recent paper.<sup>8</sup>

The sample was mounted horizontally in an oven. The oven temperature was stabilized at  $25.0 \pm 0.05^\circ\text{C}$  for 5CB samples and at  $35.0 \pm 0.05^\circ\text{C}$  for 8CB samples. The dc magnetic field  $\vec{H}$  is applied parallel to the NLC film surface. The molecular reorientation of the NLC film is probed by the optical birefringence technique: a He-Ne laser beam polarized at  $45^\circ$  to the magnetic field is normally incident to the NLC film. As the molecules rotate in the plane of  $\vec{H}$  and the normal of the NLC film surface, the refractive index of the e-ray is changed with the molecular orientational angle  $\theta$ . Thus, with an analyzer crossed with the polarizer, the output probe beam intensity,  $I_0(t)$ , is proportional to  $\sin^2 [\Delta\Phi(t)/2]$ , where  $\Delta\Phi(t)$  is the phase difference between the o-beam and e-beam after the probe beam traversing the NLC film. Using the photo detector to sense the output intensity and a Y-T recorder to record the variation of  $I_0$  with time, we can deduce the variation of the molecular reorientation angle  $\theta$  with time when the magnetic field is changed.

Figure 1 shows the variation of the relaxation rate  $1/\tau_d$  of a 5CB HB and a FS sample with respect to  $H^2$ . As the magnetic field is changed from  $H_1 > H_c$  to  $0 < H < H_c$ ,  $\tau_d$  becomes  $\tau_{\text{off}}$ , according to our definitions.

The variation of the zero field turn off rate  $1/\tau_{\text{off}}(0)$  with  $1/d^2$  for 5CB HB and FS samples are shown in Figure 2. From which we can see that the zero field turn off rate of FS samples is faster than HB samples by a factor about 1.95 for the same thickness. This is consistent with the experimental result of Reference 3, in which the molecular reorientation of the NLC film is induced by a laser light.

A set of measured differential relaxation rates as a function of  $H^2$  for 5CB HB and FS samples are presented in Figures 1(a) and 1(b), respectively.

In the range of  $0 < H < H_c$ ,  $\tau_{\text{off}}$  is equal to  $\tau_d$ , as mentioned above. From Figure

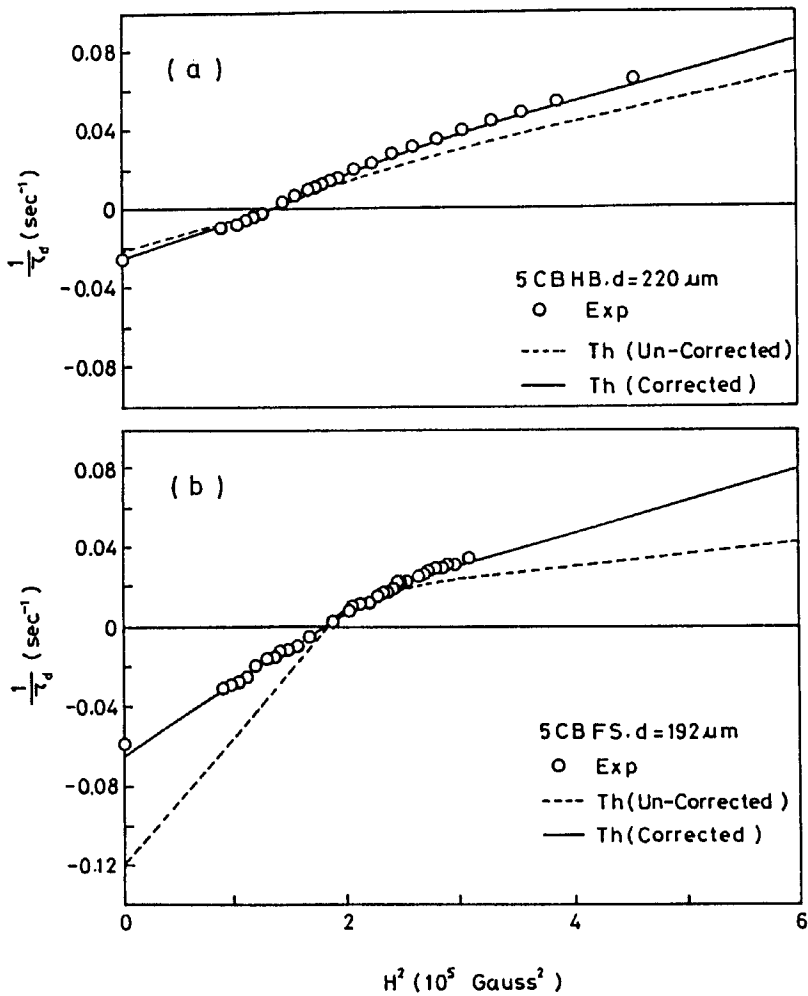


FIGURE 1 Inverse of the differential time constant versus the square of magnetic field for (a) a 220  $\mu\text{m}$  thick 5CB HB sample, and (b) a 192  $\mu\text{m}$  thick 5CB FS sample. Open circles: experimental; Dashed curves: theoretical, with viscosity coefficients from the literature; Solid curves: theoretical, with corrected viscosity coefficients.

1, one observes that  $1/\tau_{\text{off}}$  varies linearly with  $H^2$ , as predicted by the theory (see Equations 2 and 4). For 5CB, the slope of the curve of  $1/\tau_{\text{off}}$  versus  $H^2$  of the FS sample is about 1.95 times larger than that of HB sample. Thus the effective viscosity of the FS sample ( $\gamma_{1,fs}^*$ ) is about 1.95 times smaller than that of the HB sample ( $\gamma_{1,hb}^*$ ). For 8CB, the effective viscosity of FS samples are 2.0 times smaller than that of HB samples.

We have shown that the linear relation of  $1/\tau_{\text{off}}$  and  $1/\tau_d$  with  $H^2$  in the range of  $0 < H < H_c$  are in agreement with the theoretical prediction. However, the quantitative behavior of the former is very different from the latter. For example, the turn-off relaxation rate of the FS sample is about 4 times faster than the HB case,

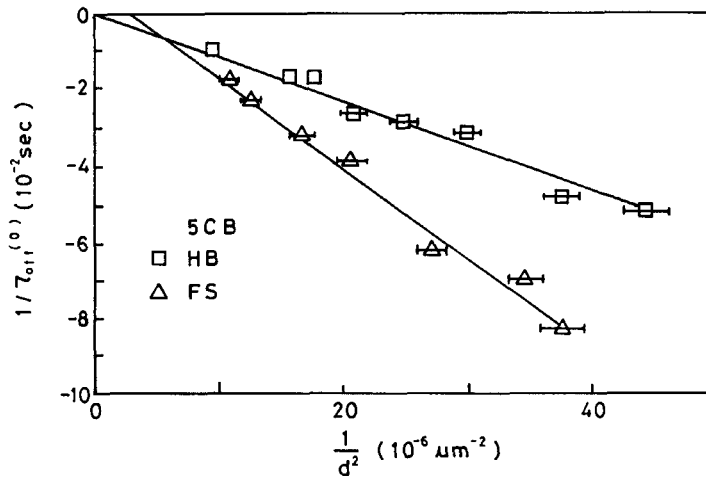


FIGURE 2 The inverse of the turn-off time constant obtained when the magnetic field is changed from  $H > H_c$  to zero as a function of  $1/d^2$  of 5CB samples.

if the parameters from literature ( $k_{11} = 0.54 \times 10^{-6}$  dyne,  $k_{33} = 0.72 \times 10^{-6}$  dyne,  $\chi_a = 1.047$  cgs units,  $\gamma_1 = 0.806 P$ ,  $\gamma_2 = -0.894 P$ ,  $\alpha_2 = -.8485P$ ,  $\alpha_3 = -.0432 P$ , and  $\eta_c = 1.12 P$ )<sup>10,11</sup> are used, but the experimental results shows 1.95 times only. Figure 1(a) is the variation of  $1/\tau_d$  with  $H^2$  for a 5CB HB sample 220  $\mu\text{m}$  in thickness. Compare the measurement data (open circles) with the theoretical prediction (dashed curve) with the above parameters, the difference is obvious. Similar results of 5CB FS sample are shown in Figure 1(b).

Results for a 8CB HB sample and a FS sample are similar to that of 5CB samples. These discrepancies in  $1/\tau_d$  can be caused by the uncertainties in the values of some of the parameters used according to the measurement error in the cited literature. The measurement principles of the previous workers are also different from ours. We can derive some of the parameters from our experimental results by comparing with the theories. From Figure 2, the slopes of the lines of the  $1/\tau_{off}(0)$  vs.  $1/d^2$  for 5CB HB samples and FS samples are found to be  $-1.2145 \times 10^{-5}$  and  $-2.373 \times 10^{-5} \text{ cm}^2/\text{sec}$ , respectively. According to Equation (5), we can write:

$$\frac{\pi^2 k_{33}}{[1 - \alpha_2^2/(6\gamma_1\eta_c)]\gamma_1} = 1.215 \times 10^{-5},$$

$$\text{and} \quad \frac{\pi^2 k_{33}}{[1 - \alpha_2^2/(\gamma_1\eta_c)]\gamma_1} = 2.37 \times 10^{-5}. \quad (8)$$

From here, we get  $\gamma_1\eta_c/\alpha_2^2 = 1.87$  cgs units. The experimental results of  $H_c$ , which is a function of  $k_{33}$ , are in good agreement with the calculated ones. We can thus assume the value of  $k_{33}$  is reliable. Because  $\gamma_1 = \alpha_3 - \alpha_2$  and  $|\alpha_3| \ll |\alpha_2|$ , the influence of  $\alpha_3$  is small. We can further assume that  $\alpha_3$  needs not to be adjusted, either. After substituting the values of  $k_{33}$  and  $\alpha_3$  obtained from the literature<sup>10,11</sup>

into Equation (8), we get  $\gamma_1 = 0.64$  (thus  $\alpha_2 = -0.683$ ) and  $\eta_c = 1.38$  cgs units. These are consistent with the results of Ru-Pin Pan *et al.*, in Reference (3).

Similarly, for 8CB, we obtain  $\gamma_1 = 0.801$  and  $\eta_c = 1.63$ . These are about twice as large as the corresponding values of A. G. Chmielewski in Reference (12). The comparison between these corrected values and the corresponding values obtained from the literature are shown in Table I.

Using these corrected parameters, the calculated theoretical values of  $1/\tau_d$  of 5CB films are shown as the solid curves in Figures 1(a) & 1(b) for 220  $\mu\text{m}$  HB sample and 192  $\mu\text{m}$  FS sample, respectively. Compare the corrected curves with the corresponding experimental data (open circles), the former is in excellent agreement with the latter for small  $H$ . It also matches better than the un-corrected curves (dashed curve) in the larger  $H$  range.

With these corrected viscosity coefficients, the theoretical differential relaxation rate as a function of  $H^2$  both for 5CB HB samples and FS samples, and also for the case of neglecting the backflow effect are calculated and shown in Figure 3. The viscosity constants  $\gamma_1$  and  $\eta_c$  deducted from several laser induced Freedericksz transition experiments<sup>3,13</sup> always show some difference from the values found in the literature. These discrepancies have been interpreted as a thermal heating effect or a laser beam size effect. From this work we would conclude that these effects are not the main reasons for the discrepancies. The turn off time constant  $\tau_{\text{off}}$  is very sensitive to the variation of  $\gamma_1$  and  $\eta_c$ . In our work,  $\gamma_1$  and  $\eta_c$  for 5CB are adjusted only by 20% and 5% from the values in References 13 and 18, respectively. This causes the turn off time constant  $\tau_{\text{off}}$  of FS sample to be doubled. Measuring the turn off time constants of HB and FS samples thus provides a sensitive method to determine the viscosity coefficients  $\gamma_1$  and  $\eta_c$ .

TABLE I

A comparison between the corrected viscosity coefficients from this work and the corresponding values obtained from the literatures, for 5CB at 25°C and for 8CB at 35°C.

|              | 5CB               |                   |           | 8CB               |           |
|--------------|-------------------|-------------------|-----------|-------------------|-----------|
|              | from<br>Ref. (11) | from<br>Ref. (12) | Corrected | from<br>Ref. (12) | Corrected |
| $\gamma_1$   | 0.806             | 0.93              | 0.64      | 0.48              | 0.801     |
| $\gamma_2$   | -0.894            | -0.93             | -0.726    | -0.48             | -0.886    |
| $\alpha_2$   | -0.849            | -0.93             | -0.683    | -0.48             | -0.843    |
| $\alpha_3^*$ | -0.043            | $\approx 0$       | -0.043    | $\approx 0$       | -0.043    |
| $\eta_c$     | 1.12              | 1.17              | 1.38      | 0.774             | 1.63      |

\*  $\alpha_3$  is not adjusted in this work.



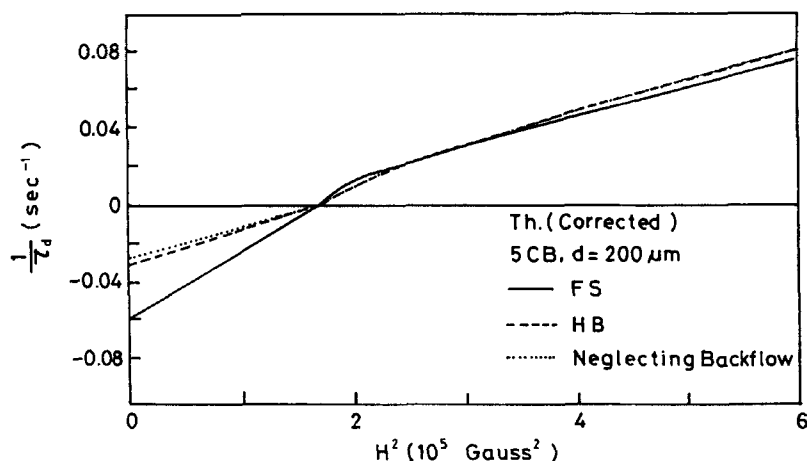


FIGURE 3 The theoretical curves of the differential relaxation rate ( $1/\tau_d$ ) versus the square of magnetic field for the 5CB films. The corrected viscosity coefficients are used.

### Acknowledgment

This work was supported by the National Science Council of the Republic of China under Grant NSC78-0208-M009-16.

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