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Free-surface effects on the response of nematic liquid crystal films to a laser pulse

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The dynamic response of 5CB films with a free surface to a laser pulse is investigated. A magnetic field above the Fréedericksz transition is applied initially to induce a starting angle on the molecular orientation. A single 1·06 μm laser pulse with e^{-1} pulse width 0·2 ms is incident normally upon the films. When the laser pulse polarization is perpendicular to the magnetic field, only a thermal effect is involved. While it is parallel to the magnetic field, both molecular orientation and thermal effects are involved. The results from films with a free surface are compared with those from films sandwiched between two glass substrates. The free surface effect, beam size effect, and thermal effect are discussed by comparing with theoretical analysis.

1. Introduction

For nematic liquid crystal (NLC) films with an air–liquid crystal interface (free surface, FS), the dynamic responses to external fields can be different from films sandwiched between two glasses, the films with so-called hard boundaries (HB). The back-flow effect (i.e. the molecular reorientation motion induces fluid-flow, then the fluid-flow inverts to influence the reorientation motion) can have a larger influence on the relaxation property of FS films, due to the relaxing of the boundary condition of the flow velocity at the free surface [1–4]. We have demonstrated the dynamic enhancements due to the free surface in Fréedericksz transitions induced either by a laser beam [3] or by a magnetic field [4].

The relaxation dynamic properties of molecular reorientation induced by an intensive laser pulse in a homeotropic NLC film with hard boundaries had been studied by Hsiung *et al.* [5]. The influence of the non local effect [6] (i.e. the reorientation motions of the molecules in the NLC film are strongly correlated) on the differential relaxation dynamic response was considered, but the influence of the back-flow effect was shown to be small and could be neglected.

In this work, the molecular reorientation induced by a single laser pulse in a homeotropic NLC film with a free surface is studied with a Nd: YAG laser. For comparison, the HB case is also studied. Both the back-flow and the non-local effects on the relaxation dynamic properties are considered in our analysis.

In § 2 the experimental method and the set-up are described. In § 3 the theoretical derivation of the relaxation dynamics of the molecular reorientation induced by the laser pulse are presented for both HB and FS cases. Particularly, we compare the back-flow and the non-local effects on the relaxation rate for both cases. The experimental results and the comparison with theory are presented in § 4. The thermal effects induced by the laser pulse are discussed in § 5. The conclusions from this work are given in § 6.

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2. Experimental methods

The experimental set-up is shown in figure 1, the NLC sample was horizontally mounted in the oven with the temperature at 25±0·05°C, A FS sample and a HB sample with thicknesses of 200 μm and 216 μm, respectively, were studied. The process of preparation of the samples has been reported elsewhere [3]. The film thicknesses were measured by an optical interferometric method [7] within an accuracy of about ± 2 μm. A static magnetic field **H** was applied parallel to the surface plane of the sample. The surface normal is along the **z** direction. A polarized single laser pulse at 1·06 μm from a Nd : YAG laser with energy 80 mJ was focused by a lens (L1) and was nearly at normal incidence on to the sample. The pump laser spot size w (e^{-2} intensity diameter) at the sample was about 500 μm.

The molecular reorientation was probed by the optical birefringence technique. A normal incidence He–Ne laser beam was polarized at 45° to the magnetic field **H** and to the polarization plane of the pump laser pulse by a polarizer (P) and then focused by a lens (L2). It overlaps with the pump laser beam at the sample. The transmitted probe

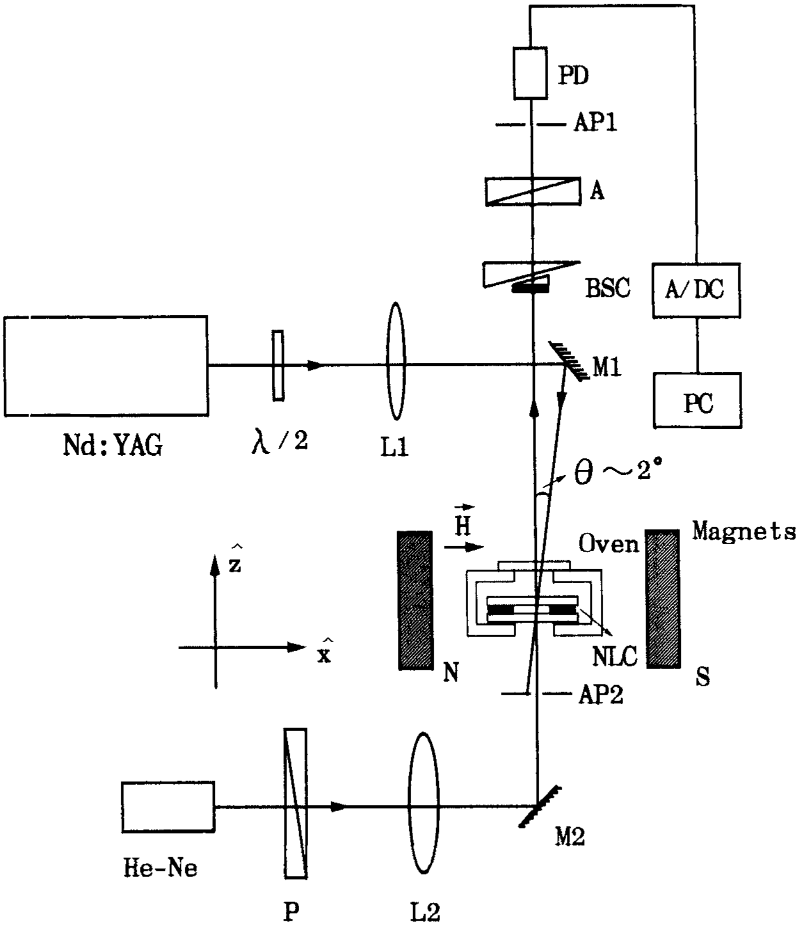


Figure 1. The experimental set-up. NLC: nematic sample; H, magnetic field; L1, L2, lenses; P, polarizer; A, analyser; SBC, Soleil–Babinet compensator; AP1, AP2, apertures; PD, photo-detector; A/DC: analogue to digital converter; PC, personal computer.

beam passed through a crossed analyser (A). A small aperture (AP1) was used such that only the central portion of the cylindrical region illuminated by the pump pulse was probed. As the molecules rotate in the **H**–**z** plane, the effective refractive index of the e ray is changed with the molecular reorientational angle θ [8]

$$n_{\mathrm{eff}}(\theta)=(n_{\mathrm{o}}^{-2}\cos^2\theta+n_{\mathrm{e}}^{-2}\sin^2\theta)^{-1/2},$$

where n_{o} and n_{e} are ordinary and extraordinary refractive indices, respectively, corresponding to the wavelength of the probe beam. The phase retardation $\Delta\phi$ between the e ray and the o ray was detected by the intensity measured at PD (photo-detector).

In the case of a near-normal-incidence cw laser beam, the onset of molecular reorientation is delayed for a long time after the laser beam is switched on [9]. Similarly, the molecules may not rotate when the short pump laser pulse is applied, because the delay time is much longer than the laser pulse duration. A static magnetic field above the Fréedericksz transition is applied to introduce an initial molecular reorientation. The reorientation in the **H**–**z** plane from the **z** direction has a spatial variation $\theta_\infty(z)$.

If the pump beam polarization is parallel to **H**, even a relatively weak pump intensity can induce an extra molecular reorientation $\Delta\theta$ in the **H**–**z** plane. The total molecular reorientation θ is

$$\theta(z,t)=\theta_\infty(z)+\Delta\theta(z,\rho,t).$$

Other than inducing molecular reorientation, the laser purse also heats the sample and thermally induces a transient change in the birefringence. The probe beam experiences optical phase change due to both the molecular reorientation effect and the thermal effect. The total phase retardation $\Delta\phi_{\mathrm{tot}}(t)$ between e ray and o ray of the probe beam through the NLC film is $\Delta\phi_{\mathrm{tot}}(t)=\Delta\phi_\infty+\Delta\phi$, where $\Delta\phi_\infty$ is due to the static molecular reorientation angle θ_∞, and $\Delta\phi(t)$, the transient part of phase retardation is

$$\phi(t)=\Delta\phi_{\parallel}(t)=\Delta\phi_{\mathrm{mr}}+\Delta\phi_{\mathrm{th}}, \qquad (1\,a)$$

where

$$\Delta\phi_{\mathrm{mr}}(t)=\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\frac{\partial}{\partial\theta}[n_{\mathrm{eff}}(\theta)]_{\theta_\infty}\Delta\theta(z,t)\,dz, \qquad (1\,b)$$

and

$$\Delta\phi_{\mathrm{th}}(t)=\frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\frac{\partial}{\partial T}[n_{\mathrm{eff}}(\theta)-n_{\mathrm{O}}]_{\theta_\infty}\Delta T(z,t)\,dz, \qquad (1\,c)$$

where $\Delta\phi_{\mathrm{mr}}$ and $\Delta\phi_{\mathrm{th}}$ are the optical phase differences due to the laser pulse induced molecular reorientation and the thermal effect, respectively.

To detect the small change $\Delta\phi(t)$, a Soleil–Babinet compensator (SBC) was used between the sample and the analyser, and adjusted such that the probe beam output intensity was

$$I_{\mathrm{o}}(t)=(I_{\mathrm{i}}/2)\{1\pm\sin[\Delta\phi(t)]\}. \qquad (2)$$

The output intensity of the probe beam was detected by a photo-detector (PD), connected to an A/D converter and then to a personal computer to record the variation of I_{o} with time. Then, $\Delta\phi(t)$ was deduced.

If the pump beam was polarized perpendicular to the **H**–**z** plane, the molecular reorientation by the pump beam was negligibly small; the phase change is caused by laser heating only, i.e.

$$\Delta\phi(t)=\Delta\phi_{\perp}(t)=\Delta\phi_{\mathrm{th}}.$$

The polarization of the pump beam was changed with a half-wave plate placed after the laser output mirror.

In the liquid crystal, the thermal relaxation time constant is generally much shorter than the orientational relaxation time [5]. Therefore, $\Delta\phi_{\mathrm{mr}}(t)$ could be obtained simply by subtracting $\Delta\phi_{\perp}(t)$ from $\Delta\phi_{\parallel}(t)$.

In order to avoid fast thermal expansion on the free surface of the FS sample, the Nd:YAG laser was not Q-switched and was operated in the single-shot mode. The time duration of the laser pulse (e^{-1}) was about 0·2 ms.

3. Theory

With a constant magnetic field **H** greater than the Fréedericksz transition threshold $\mathbf{H}_c$, the liquid crystal molecules reorient from the **z** direction toward **H** with angle $\theta_\infty(z)$. When a laser pulse is further applied to the NLC film, the molecules will rotate through an additional angle $\Delta\theta$. After the pulse, the molecular orientation will then relax back to its steady state.

The above relaxation time constant is similar to the differential time constant of molecular reorientation, τ_d, induced by a small change in magnetic field about a constant magnetic field [2, 4]. Thus the theoretical analysis for the small variation in magnetic field [4] can be applied here. However, two major differences occur. One is the thermal effect as mentioned above. Another is the non-local effect caused by the finite beam size of the pump laser. The reorientation is restricted in a small region around the laser beam, while it is uniform under a uniform magnetic field.

Here, we use a similar procedure as in [4], but incorporating the non-local effect to derive the differential dynamic relaxation properties induced by a laser-pulse.

When the laser pulse is over, the coupled differential equations of motion for the molecular reorientation and the fluid-flow velocity under magnetic field **H** are [4, 10]:

$$\gamma_1\frac{\partial\theta}{\partial t}=[K_{33}+(K_{11}-K_{33})\sin^2\theta]\left(\frac{\partial^2}{\partial z^2}+\nabla_t^2\right)\theta+\tfrac{1}{2}(K_{11}-K_{33})\sin(2\theta)\left[\frac{\partial\theta}{\partial z}\right]^2$$
$$+\tfrac{1}{2}\chi_a H^2\sin(2\theta)+\tfrac{1}{2}[\gamma_1-\gamma_2\cos(2\theta)]\frac{\partial v_x}{\partial z}, \qquad (3a)$$

and

$$\frac{\partial}{\partial z}\left[(\alpha_1\sin^2\theta\cos^2\theta-\tfrac{1}{2}\alpha_2\cos^2\theta+\tfrac{1}{2}\alpha_3\sin^2\theta+\tfrac{1}{2}\alpha_4+\tfrac{1}{2}\alpha_5\cos^2\theta\right.$$
$$\left.+\tfrac{1}{2}\alpha_6\sin^2\theta)\frac{\partial v_x}{\partial z}+(\alpha_2\cos^2\theta-\alpha_3\sin^2\theta)\frac{\partial\theta}{\partial t}\right]=0. \qquad (3b)$$

where K_{11}, K_{33} are Frank elastic constants, and $\chi_a\equiv\chi_{\parallel}-\chi_{\perp}$, with $\chi_{\parallel}$ and $\chi_{\perp}$ being the diamagnetic susceptibilities parallel and perpendicular to the director, respectively; $\gamma_1\equiv\alpha_3-\alpha_2$, $\gamma_2\equiv\alpha_3+\alpha_2$, and $\alpha_1,\ldots,\alpha_6$ are the Leslie's viscosity coefficients following the notations in [11]. Here, the term with transverse coordinate Laplacian, $\nabla_t^2\theta$, is included to reflect the non-local effect [10]. In general, equation (3 *b*) should also include a term with $\nabla_t^2 v_x$ [10]. However, it is neglected, because either the back-flow effect or the non-local effect is a first order correction to the dynamic response, and the $\nabla_t^2 v_x$ is a second order correction term. For a laser beam with a radially symmetric intensity profile, we can assume the molecular reorientation angle $\Delta\theta$ induced by the pump laser is also a radially symmetric function. In the cylindrical coordinate, the transverse laplacian operator becomes

$$\nabla_t^2 = \nabla_\rho^2 = \frac{\partial^2}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial}{\partial\rho}. \tag{4}$$

The same procedure, boundary conditions at $z = \pm d/2$ and trial solutions, as given in [4] for a uniform magnetic field are used here, except that the small variation of the molecular reorientation $\Delta\theta$ due to the laser pulse is now also ρ dependent. Let $\Delta\theta(\rho, z, t) = \phi_m(\rho, t)\cos(\pi z/d)$; these equations of motion reduce to

$$\dot{\phi}_m + \frac{1}{\tau_{dL,i}}\phi_m = \frac{C}{D_i}\nabla_\rho^2\phi_m, \tag{5}$$

where i = HB or FS. The parameters $C = (1/\gamma_1)[K_{33} + \frac{1}{4}(K_{11} - K_{33})(1 + G_2)]$, and $D_i = 1 + (B/A_i)(\gamma_i/\gamma_1)$, and G_2, γ_i are derived and defined in [1]. The subscript L is for laser.

By separation of the variable method and letting $\phi_m(\rho, t) = R(\rho)T(t)$, we obtain the following from equation (5),

$$\dot{T} + \left(\frac{1}{\tau_{dL,i}} + \frac{1}{\Gamma}\right)T = 0, \tag{6a}$$

and

$$\frac{C}{D_i}\nabla_\rho^2 R + \frac{1}{\Gamma}R = 0, \tag{6b}$$

where Γ is a constant. With the simplified boundary condition $\phi_m(\rho \geqslant w) = 0$, the general solutions of equation (6b) are zeroth order Bessel functions, $J_0(S_n\rho/w)$, for $n = 1, 2, \ldots$, where S_n is the nth root of J_0. That is

$$\frac{1}{\Gamma} = \frac{C}{D_i}(S_n/w)^2 = \frac{1}{\Gamma_n}. \tag{7}$$

Substituting equation (7) into equation (6a) we obtain the differential rate as

$$\frac{1}{\tau_{dL,i,n}} = \frac{1}{D_i}\left[\frac{1}{\tau_{do}} + C(S_n/w)^2\right], \tag{8}$$

where, $\tau_{dL,i,n}$ is the nth mode differential time constant caused by a laser pulse, for i = HB or FS. The general solution of equation (5) is given by

$$\phi_{m,i}(\rho, t) = R(\rho)T(t) = \sum_{n=1}^{\infty} a_{i,n}J_0(S_n\rho/w)\exp(-t/\tau_{dL,i,n}), \tag{9}$$

where the $a_{i,n}$s are coefficients. We assume that the first term dominates; the differential relaxation time constant is now given by equation (8) with $n = 1$ and $S_1 = 2{\cdot}405$.

In summary, we compare the influences of the back-flow and the non-local effects on the relaxation dynamic properties of molecular reorientation induced by the laser-pulse as follows:

(1) if we neglect both the back-flow and non-local effects, i.e. $B = C = 0$, then $\tau_d = \tau_{d_o}$ for both HB and FS [4];

(2) if we consider the back-flow effect but neglect the non-local effect, i.e. $C = 0$, then $\tau_d = \tau_{d,i}$ as in the uniform field cases, where i = HB or FS [4];

(3) if we consider the non-local effect but neglect the back-flow effect, i.e. $B = 0$ and $(D_i = 1)$, then

$$\tau_d = \left[\frac{1}{\tau_{do}} + C(S_1/w)^2\right]^{-1}$$

for both HB and FS;

(4) if we keep both the back-flow and non-local effects, the relaxation time constant,

$$\tau_d = {}_{\mathrm{dL},i} = D_i\left[\frac{1}{\tau_{do}} + C(S_1/w)^2\right]^{-1} = \left[\frac{1}{\tau_{d,i}} + \frac{C}{D_i}(S_1/w)^2\right]^{-1},$$

where i = HB or FS.

The calculated results for $1/\tau_d$ as functions of H^2 for these four situations are shown in figure 2 for HB and FS samples with $d = 200\ \mu$m. The parameters used for 5CB are the same as those used in [4]: $K_{11} = 0{\cdot}54 \times 10^{-6}$ dyne, $K_{33} = 0{\cdot}72 \times 10^{-6}$ dyne, $\chi_a = 1{\cdot}047$

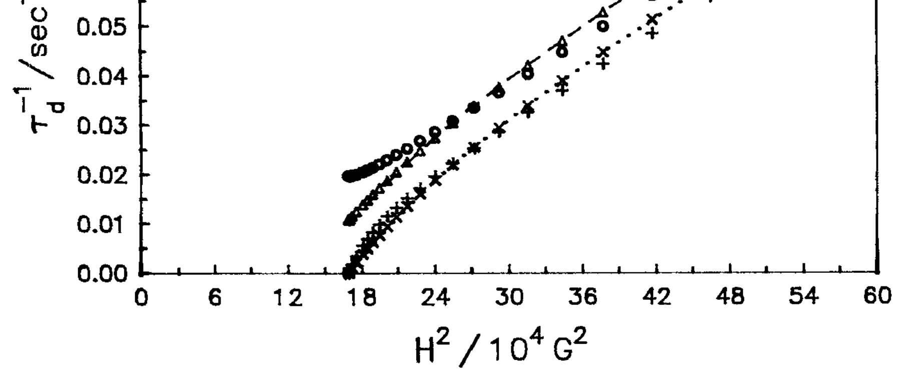


Figure 2. The theoretical curves of the differential relaxation rate $(1/\tau_d)$ of a 200 μm thick film versus the square of the magnetic field (H^2), for: (...) HB and FS cases, neglect both back-flow effect (BFE) and non-local effect (NLE); (×) HB case, keep BFE, neglect NLE; (+) FS case, keep BFE, neglect NLE; (- - -) HB and FS cases, keep NLE, neglect BFE; (△) HB case, keep BFE and NLE; and (○) FS case, keep BFE and NLE.

10^{-7} cgs units [12], and the adjusted viscosity coefficients: $\alpha_1 = 0{\cdot}08$ P, $\gamma_1 = 0{\cdot}68$ P, $\gamma_2 = -0{\cdot}726$ P and $\eta_c = 1{\cdot}53$ P. From these curves, it is shown that the back-flow effect decreases the differential relaxation rate slightly for the FS film, but can be completely neglected for the HB film (these justify the results of Hsiung and *et al.* [5]). On the other hand, the influence of the non-local effect increases the relaxation rate for both HB and FS films. The relaxation rates of the HB and FS films with the same thickness are only slightly different due to the back-flow effect.

4. Experimental results and comparison with theories

Typical traces of the measured phase retardations *vs* time for pump pulse polarization parallel and perpendicular to the H–plane (i.e. $\Delta\phi_{\parallel}$ and $\Delta\phi_{\perp}$, respectively), and the difference between these two (i.e. $\Delta\phi_{\mathrm{mr}}$), are shown in figure 3 (*a*) for the HB sample under a magnetic field ($H = 735$ Gauss). It shows that $\Delta\phi_{\mathrm{mr}}$ can be fitted with an exponential function (shown as a solid curve in the figure) very well. The measured orientational relaxation time is obtained from the exponential time constant of this fitting. Similar results for the FS sample are shown in figure 3 (*b*).

The relaxation rates ($1/\tau_d$) determined in this way under different magnetic fields are then plotted against H^2 with solid circles in Figure 4 (*a*) and (*b*), for the HB and FS samples, respectively.

In order to study the non-local effect on the relaxation dynamic properties, the reorientation relaxation rates induced by differential variation of the magnetic field around a constant field **H** are also measured and shown in figure 4 with crosses. The theoretical values are shown in the figures together with the experimental data. The dotted curve is from calculation, considering the back-flow effect, but not the non-local effect, it gives a perfect fit for the differential time constant by varying the magnetic field slightly. The perfect match has been discussed in [4]. The solid curve is calculated with the back-flow and the non-local effect caused by finite laser beam size. The theoretical calculations predict that the relaxation rate of the molecular reorientation induced by the laser pulse is about $0{\cdot}008\ \mathrm{s}^{-1}$ faster than that by changing the magnetic field for both HB and FS samples in our experimental magnetic field range. This increase due to the non-local effect is very close to our measured relaxation rate increase, comparing with the relaxation rate under a constant magnetic field.

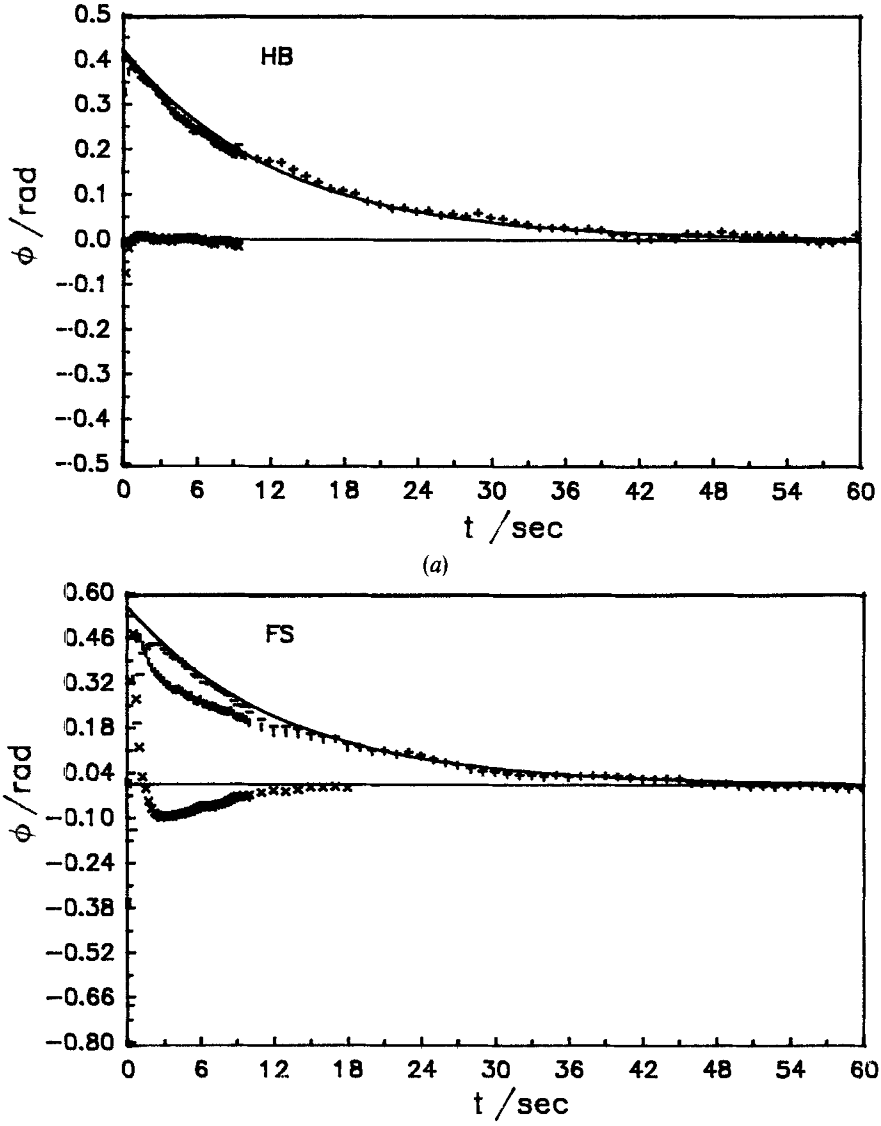
5. The thermal effect

The thermal relaxation time is found to be shorter than the orientational relaxation time for both the HB and FS cases. However, it is much shorter in the FS case than the HB case.

Since the pump beam diameter was much larger than the sample thickness in our experiment, we can neglect heat diffusion in the transverse plane. The temperature change ΔT then obeys the one-dimensional heat-diffusion equation

$$\frac{\partial}{\partial t}\Delta T = \alpha \frac{\partial^2}{\partial z^2}\Delta T. \tag{10}$$

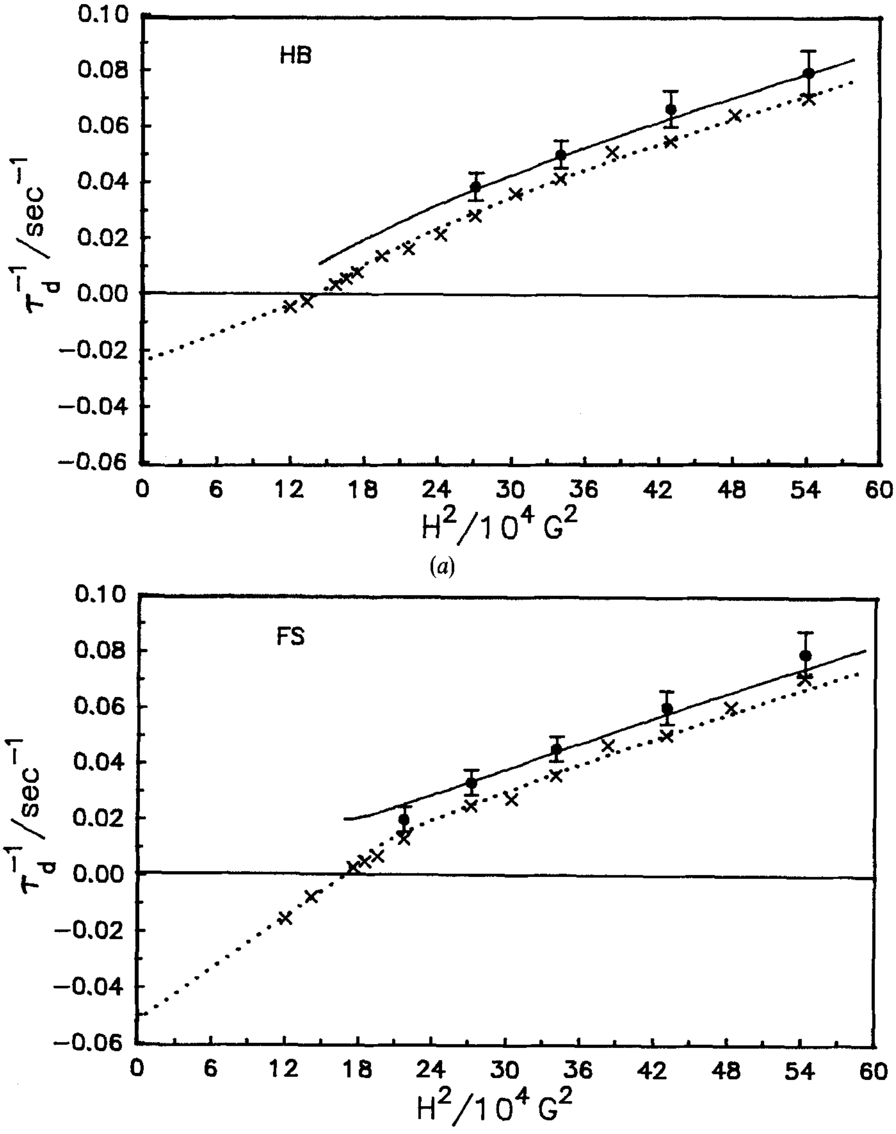
We have assumed here that the thermal diffusivity α is independent of the director orientation in the medium. For the HB film, the glass of the sample can be considered as a good heat sink because of its good heat conductivity, compared to air and liquid crystal.



(*a*)

(*b*)

Figure 3. The phase retardation of the probe beam induced by a 80 mJ laser pulse as a function of time for (*a*) a HB sample with $d = 216\,\mu$m, and (*b*) a FS sample with $d = 200\,\mu$m. (+) $\Delta\phi_{\parallel}(t)$, measured with the pump polarization parallel to **H**; (×) $\Delta\phi_{\perp}(t)$, measured with the pump polarization perpendicular to **H**; (−) $\Delta\phi_{mr} = \Delta\phi_{\parallel} - \Delta\phi_{\perp}$; and (——) the fitted curve $\Delta\phi_{mr}(t) = \Delta\phi_0 \exp(-t/\tau_d)$.



(*a*)

(*b*)

Figure 4. The relaxation rate versus the square of magnetic field for (*a*) HB sample with $d = 216\,\mu$m and (*b*) FS sample with $d = 200\,\mu$m. (●) induced by the laser pulse; (×) induced by the small change in magnetic field; Solid curve: theoretical, keeping the BFE and NLE; dotted curve: theoretical, keeping the BFE, but neglecting the NLE.

The boundary conditions for the HB sample can be set as $\Delta T(z=\pm d/2)=0$ [5]; the general solution of equation (10) is

$$\Delta T(z,t)=\frac{4\Delta T_0}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1}\cos\left[(2n+1)\frac{\pi z}{d}\right]\exp(-t/t_n),$$

where $1/t_n=\alpha(2n+1)^2(\pi/d)^2$, and $\Delta T_0=\Delta T(t=0^+)$. In general, the t_n are so small that only the first two modes, t_0 and t_1, are sufficient, as shown in the experimental results (see figure 2 (*a*)).

For FS sample, because the heat conductivity of the air is very poor, the boundary condition of ΔT at the free surface no longer equals 0. If we assume that the boundary condition at the free surface is $\partial(\Delta T)/\partial z=0$, the solution of equation (10) has the form of

$$\Delta T(z,t)=\sum_{n=0}^{\infty}c_n\sin\left[(2n+1)\frac{\pi z}{2d}\right]\exp(-t/t_n),$$

where $1/t_n=\alpha(2n+1)^2(\pi/2d)^2$, the c_ns are coefficients, and we have redefined the origin point of the z axis at the hard-boundary of the FS sample. Thus, the thermal relaxation time constants of the FS case are four times those of the HB case, for each corresponding mode. However, the measured thermal relaxation time of the FS case is far more than four times larger. Thus more studies are required on this problem.

6. Conclusion

The differential relaxation dynamic properties of the molecular reorientation induced by a laser pulse in nematic liquid crystal films have been measured and derived, for both the FS and the HB cases. The influences of the back-flow and the non-local effects on the relaxation rates are compared. We have found that the influence of the non-local effect is larger than that of the back-flow effect for both cases, when a static magnetic fixed greater than H_c is present. In general, the difference between the FS and the HB films with the same thickness is small, and is within experimental error.

We have also compared theoretically the relaxation rates of the molecular reorientation induced by the laser pulse and by a small change in uniform magnetic field under the same constant magnetic field, and by consideration of the non-local effect have found that the former is faster than the latter by about $0{\cdot}008\ \mathrm{s}^{-1}$, for both the HB and FS films of about 200 μm thickness. Our experimental measurements agree very well with our prediction.

The relaxation time of the thermal effect is found to be smaller than the reorientation relaxation time for both the HB and FS cases. However, the thermal relaxation time of the FS film is much larger than that of the HB case.

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