Magnetic-field-enhanced and -suppressed intrinsic optical bistability in nematic liquid crystals

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It is shown that the optical-field-induced first-order molecular reorientation and intrinsic optical bistability in a nematic liquid crystal can always be enhanced or suppressed by a low magnetic field (typically < 1 kG), and hence can be seen in all existing nematics at a low optical power (typically < 1 kW/cm^2).

The study of optical nonlinearities via molecular reorientation of liquid crystals (LC's) is of particular interest because among fluids, LC's have the largest optical-field-induced refractive index changes. The reorienting torque produced by a cw optical field on LC's can result in an extremely strong collective molecular reorientation and large associated nonlinear effects.[1] For example, the light, self-focusing nonlinear constant of nematic LC's (NLC's) has a value larger by eight to ten orders of magnitude than that of carbon disulfide. Recently, we obtained the exact solution for describing the NLC orientation, and found that certain existing NLC's can have first-order and optical bistable (OB) transitions, in which increasing optical field intensity results in discontinuous changes in the LC orientation.[2,3] This form of intrinsic OB[4] does not use a resonant optical cavity, and offers the attractive possibility of making large electroresponse devices with a low-laser–power sharp threshold.

Our early study showed that, without an additional magnetic field, the criterion for the existence of the intrinsic optical bistability in NLC's is given by $k_{11}/k_{33}+9n_0^2/4n_e^2<\frac{9}{4}$, where k_{11} and k_{33} are the splay and bend elastic constants, and n_0 and n_e are the ordinary and extraordinary indexes of refraction.[2,3] This OB criterion can be satisfied by the following four room-temperature NLC mixtures: RO-TN-200, RO-TN-201, RO-TN-403 (from Hoffmann–La Roche), and *E*7 (from British Drug House), and a high-temperature single-component nematic PAA (*p*-azoxyanisole). Without an external field, the OB criterion depends completely on the NLC's material parameters. However, this study shows that the application of an additional magnetic field parallel to the laser propagation can be a significant control parameter for enhancement of OB. First we obtain the exact solution for the external field effects on the OB in NLC's. We then obtain the OB criterion and show that OB can always be enhanced or suppressed by an external magnetic field (typically < 1 kG), and hence can be seen in all existing nematics at a low-laser power (typically < 1 kW/cm^2).

We consider a NLC cell of thickness d confined between the planes $z=0$ and $z=d$ of a Cartesian coordinate system (Fig. 1). The surfaces are treated to give homeotropic alignment, i.e., NLC's are aligned parallel to the z axis. An orienting laser beam of intensity I is normally incident on the cell, with the polarization parallel to the plane of incidence, which is the xz plane. The optical field will apply a torque on the NLC so as to reorient the NLC along the x axis. We shall first consider geometry I [Fig. 1(a)], in which the additional magnetic field is directed along the z axis, $\mathbf{H}=(0,0,H)$. Known thermotropic NLC's are diamagnetic with positive diamagnetic anisotropy χ_a, so that the applied magnetic field always tends to align the director along the field direction.

The NLC's orientation can be described by the director $\mathbf{n}(z)=(\sin\theta,0,\cos\theta)$, where $\theta(z)$ is the angle between the director and the z axis. By the symmetry of the problem, we look for solutions which are symmetrical with respect to the $z=d/2$ plane, i.e., $\theta(z)=\theta(d-z)$. Variation of the total free energy (consisting of the elastic deformation energy, optical field energy, and magnetic field energy) leads to a Euler equation from which we obtain the followng solution for the director:[5]

$$z=\left[\frac{ck_{33}}{2I}\right]^{1/2}\times\int_0^\theta\left[\frac{1-k\sin^2\theta}{n_p(\theta_m)-n_p(\theta)-R[P(\theta_m)-P(\theta)]}\right]^{1/2}d\theta\ , \quad (1)$$

for $d\theta/dz\neq 0$ and $0\leq z\leq d/2$, where

$$\theta_m=\theta(z=d/2),\quad R=\chi_a H^2/2I\ ,$$

$$n_p(\theta)=n_0/\sqrt{1-u\sin^2\theta},\quad P(\theta)=\sin^2\theta\ ,$$

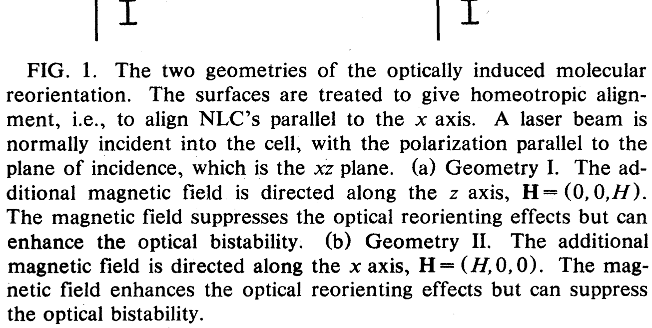

FIG. 1. The two geometries of the optically induced molecular reorientation. The surfaces are treated to give homeotropic alignment, i.e., to align NLC's parallel to the x axis. A laser beam is normally incident into the cell, with the polarization parallel to the plane of incidence, which is the xz plane. (a) Geometry I. The additional magnetic field is directed along the z axis, $\mathbf{H}=(0,0,H)$. The magnetic field suppresses the optical reorienting effects but can enhance the optical bistability. (b) Geometry II. The additional magnetic field is directed along the x axis, $\mathbf{H}=(H,0,0)$. The magnetic field enhances the optical reorienting effects but can suppress the optical bistability.

and $u=1-n_0^2/n_e^2$. $u>0$, since for all known NLC's, $n_e>n_0$. The maximum deformation angle $\theta_m=\theta(z=d/2)$ can be determined by evaluating Eq. (1) at $z=d/2$, and setting the upper limit in the integration to be $\theta=\theta_m$. The spatial orientation of the NLC at a given optical intensity I and magnetic field strength H is then completely described by Eq. (1).

In the rising transition, there exists a rising threshold intensity $I_{\rm th}$ below which no molecular reorientation can be induced. By expanding the solution for the director up to and including terms of the order of θ_m^2, we obtain that, in the rising transition, $\theta(z)=0$ for $I<I_{\rm th}$, and for $I\geqslant I_{\rm th}$.

$$\theta(z)\cong\theta_m(I)\sin(\pi z/d)+O(\theta_m^3)\ , \tag{2}$$

and

$$\theta_m(I)\cong\sqrt{(I/I_{\rm th}-1)/2B}\ , \tag{3}$$

where

$$B=B_0-9ur^2/16,\quad I_{\rm th}=I_0(1+r^2)\ ,$$

$$B_0=(1-k-9u/4)/4,\quad I_0=(ck_{33}/n_0u)(\pi/d)^2\ ,$$

$$r=H/H_0,\quad H_0=(\pi/d)\sqrt{k_{33}/\chi_a}\ ,$$

and $O(\theta_m^3)$ means terms of order of θ_m^3.

Equation (3) shows that if $B>0$, the transition is a second-order transition, in which $\theta_m=0$ for $I<I_{\rm th}$, and θ_m changes continuously as $I\geqslant I_{\rm th}$. But if $B<0$, small distortion is not stable and a first-order transition (OB) occurs. By computing the integral in Eq. (1) up to and including terms $\sim O(\theta_m^4)$, we find that

$$\theta_m(I)\cong\left[\frac{-B+(B^2+4GA)^{1/2}}{2G}\right]^{1/2}\ , \tag{4}$$

where $A=\sqrt{I/I_{\rm th}}-1$,

$$G=G_0+[(9u/4+63ku/4+639u^2/16)r^2+1539u^2r^4/32]/96\ ,$$

and

$$G_0=(11/2-k+9u/4+63ku/4-9k^2/2-261u^2/32)/96\ .$$

Equations (3) and (4) show that,[2,3] for a first-order transtion, as I increases from zero, $\theta_m=0$ for $I<I_{\rm th}$, and the state changes discontinuously at $I_{\rm th}$ from $\theta_m=0$ to $\theta_m=\sqrt{-B/G}$, and then changes continuously as I increases. But if I decreases from above $I_{\rm th}$, the state assumes a finite amount of distortion even at $I<I_{\rm th}$. Upon reaching a lower falling threshold intensity $I'_{\rm th}=I_{\rm th}(1-B^2/4G)^2$, the state changes discontinuously from $\theta_m=\sqrt{-B/2G}$ back to $\theta_m=0$. The changes at both the rising and falling threshold intensities are discontinuous, and the rising and falling transitions assume the same deformation for $I<I'_{\rm th}$ and $I>I_{\rm th}$ but different deformation for $I'_{\rm th}<I<I_{\rm th}$; we thus obtain OB. There is a tricritical point separating the second- and first-order transitions at $B=0$ at the reduced field $r^*=H^*/H_0=\sqrt{16B_0/9u}$. Here the transition is second order with $\theta_m=[(I/I_{\rm th}-1)/2G]^{1/4}$.

By interpreting θ_m as an order parameter, the total free energy F can be expanded in the form

$$F=-A\theta_m^2+B\theta_m^4+G\theta_m^6+O(\theta_m^8)\ ,$$

which is the same form assumed in the Landau theory of phase transitions. It can then be shown that the thermodynamic description of OB agrees with the above prediction on OB using the solution for the director, as we have shown in Ref. 2 for the case of $H=0$, i.e., a purely optically induced first-order transition. Thus, we conclude that, at the rising threshold, the intensity dependence of θ_m is mean-field like with the critical exponent $\frac{1}{2}$ for $B>0$ and $\frac{1}{4}$ at the tricritical point $B=0$.

The above discussion shows that the application of an additional magnetic field parallel to the laser propagation direction is a significant control parameter for the enhancement of OB. If $B=B_0-9ur^2/16<0$, OB will occur. For $B_0\geqslant 0$, the second-order transition will become a first-order transition when $H>H^*$. Thus, with a magnetic field directed along the z axis, OB can always be enhanced and can always be seen for all existing nematics. Because the magnetic field suppresses the optical orienting effects, $I_{\rm th}/I_0-1$ increases quadratically as r is increased.

We now consider geometry II in which the applied magnetic field is directed along the x axis, $\mathbf{H}=(H,0,0)$ [Fig. 1(b)]. All the above analyses for geometry I hold for this case with the replacements

$$P(\theta)=\sin^2\theta\rightarrow P(\theta)=-\sin^2\theta\ \text{ and }\ r^2\rightarrow -r^2\ .$$

A comparison between the two geometries is given in Table I. In the second geometry, the magnetic field enhances the optical orienting effects, $I_{\rm th}/I_0-1$ decreases quadratically as r is increased. If $B=B_0+9ur^2/16<0$, OB will occur. The

TABLE I. A comparison of the different parameters in geometries I and II. In the table, $k=1-k_{11}/k_{11}$, $u=1-n_0^2/n_e$, $B_0=(1-k-9u/4)/4$, $G_0=(\frac{11}{2}-k+9u/4+63ku/4-9k^2/2-261u^2/32)/96$, $r=H/H_0$, and $H_0=(\pi/d)\sqrt{k_{33}/\chi_a}$.

Parameter	$H=0$	Geometry I	Geometry II
$\mathbf{H}$	0	$(0,0,H)$	$(H,0,0)$
P	0	$\sin^2\theta$	$-\sin^2\theta$
$I_{\rm th}$	I_0	$I_0(1+r^2)$	$I_0(1-r^2)$
B	B_0	$B_0-9ur^2/16$	$B_0+9ur^2/16$
G	G_0	$G_0+[(9u/4+63ku/4+639u^2/16)r^2$ $+1539u^2r^4/32]/96$	$G_0+[-(9u/4+63ku/4+639u^2/16)r^2$ $+1539u^2r^4/32]/96$
H^*		$H_0\sqrt{16B_0/9u}$ for $B_0\geqslant 0$	$H_0\sqrt{16\lvert B_0\rvert/9u}$ for $B_0<0$

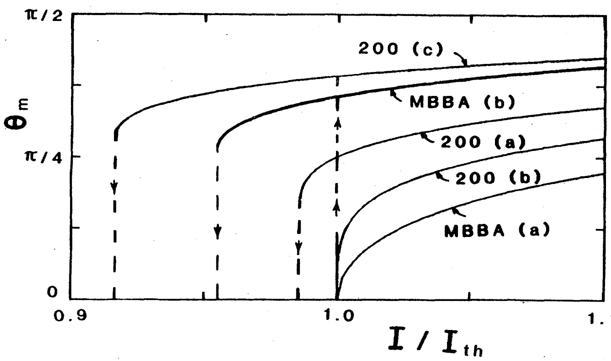

FIG. 2. Maximum deformation angle θ_m as a function of the reduced intensity I/I_{th} and magnetic field H/H_0 for RO-TN-200 and MBBA at temperature 22 °C and wavelength 6328 Å. For RO-TN-200 (abbreviation: 200) we put $k_{11}=8.80\times10^{-7}$ dyn, $k_{33}=19.00\times10^{-7}$ dyn, $n_0=1.5345$, $n_e=1.8100$, and $d=250$ μm. (a) $H=0$. The transition is first order, i.e., optical bistable with $I_{th}=I_0=208.6$ W/cm^2, $I'_{th}=0.986I_{th}=205.7$ W/cm^2. (b) $\mathbf{H}=(H^*,0,0)$. $H^*=0.518H_0$ is the tricritical field. The OB is suppressed and the transition becomes second order with $I_{th}=0.732I_0=152.6$ W/cm^2. (c) $\mathbf{H}=(0,0,H_0)$. The OB cycle is enhanced with $I_{th}=2I_0=417.1$ W/cm^2, $I'_{th}=0.917I_{th}=382.5$ W/cm^2. For MBBA we put $k_{11}=6.95\times10^{-7}$ dyn, $k_{33}=8.99\times10^{-7}$ dyn, $n_0=1.5443$, $n_e=1.7582$, $x_a=10^{-7}$ cgs unit, and $d=250$ μm. (a) $H=0$. The transition is second order with $I_{th}=I_0=120.7$ W/cm^2. (b) $\mathbf{H}=(0,0,1.5H_0)$. The OB is enhanced with $I_{th}=3.25I_0=393.2$ W/cm^2, and $I'_{th}=0.954I_{th}=374.1$ W/cm^2. $H_0=380$ G. The transition becomes first order in the field $\mathbf{H}=(0,0,H)$ with $H>H^*=0.71H_0$.

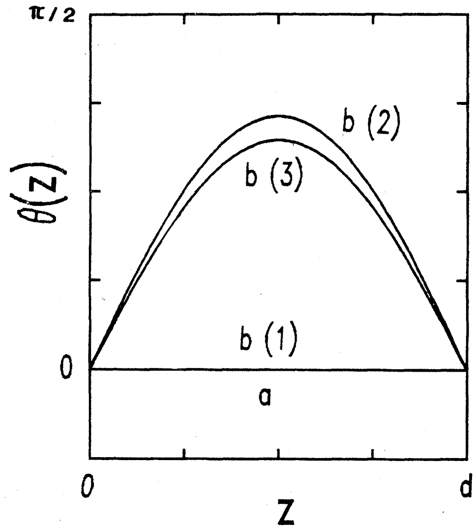

FIG. 3. Spatial bistable orientation of the deformation angle of the director in the magnetic-field-enhanced optical bistability for MBBA. (a) $H=0$. The transition is second order and $\theta(z)=0$ for all $I\le I_{th}$. (b) $\mathbf{H}=(0,0,1.5H_0)$. The transition is first order and magnetic-field-enhanced optically bistable orientation occurs. (1) In the rising transition, $\theta(z)=0$ for all $I<I_{th}$. (2) In the rising transition, deformation occurs at $I=I_{th}$, and the state changes discontinuously from $\theta_m=0$ to $\theta_m=\sqrt{-B/G}$. (3) In the falling transition starting from $I\ge I_{th}$, the state assumes a finite deformation at $I=0.97I_{th}$. Only after $I<I'_{th}=0.954I_{th}$ will the state change discontinuously from $\theta_m=\sqrt{-B/2G}$ to $\theta_m=0$.

magnetic field can suppress the OB since $B\le B_0$. If $B_0<0$, the first-order transition will become second order when $H\ge H^*=H_0\sqrt{16|B_0|/9u}$. Thus, with a magnetic field directed along the x axis, OB can always be suppressed and the threshold intensity will be reduced.

Experimentally, the purely optical-field-induced molecular reorientation has been observed in a few well-known NLC's.[1] Since all the NLC's used in the experiments have positive B_0, OB was not observed as expected. The theoretical prediction that OB could occur in NLC's (such as the four room-temperature NLC mixtures: RO-TN-200, RO-TN-201, RO-TN-403, and $E7$) satisfying $B_0<0$ (i.e., $k_{11}/k_{33}+9n_0^2/4n_e^2<\frac{9}{4}$), has not been verified experimentally.[2] This study shows that OB can always be enhanced or suppressed and hence can always be seen for all existing NLC's. This is illustrated in Fig. 2 for RO-TN-200 and MBBA [p-(n-methoxy)benzilidene-(p-butyl)aniline].[6] For RO-TN-200, $B_0<0$, and OB could occur even without an external field as shown in Fig. 2, 200 (a). The OB will be suppressed and the first-order transition becomes a second-order transition in a magnetic field $\mathbf{H}=(H,0,0)$ with $H\ge H^*=H_0\sqrt{16|B_0|/9u}=0.518H_0$ [Fig. 2, 200 (b)]. With $\mathbf{H}=(0,0,H)$, the width of the OB cycle will always be enlarged as shown in Fig. 2, 200 (c) for $H=H_0$; the falling threshold intensity becomes $I'_{th}=0.917I_{th}$ (and width $I_{th}-I'_{th}=34.6$ W/cm^2 for $d=250$ μm), as compared to $I'_{th}=0.986I_{th}$ (and width $I_{th}-I'_{th}=2.9$ W/cm^2 for $d=250$ μm) at $H=0$. For MBBA, without an external magnetic field, $B_0>0$ and OB cannot occur, as observed [Fig. 2, MBBA (a)].[2] However, OB can always be enhanced, and the second-order transition will become a first-order transition in a magnetic field $\mathbf{H}=(0,0,H)$, with $H>H^*=H_0\sqrt{16B_0/9u}=0.71H_0$, as shown in Fig. 2. MBBA (b) for $H=2H_0$. The spatial bistable orientation of the deformation angle $\theta(z)$ of the director as a function of the optical-field intensity and magnetic field strength for MBBA is shown in Fig. 3. The results show that OB is enhanced in MBBA by a magnetic field $\mathbf{H}=(0,0,1.5H_0)$. Thus, using a low magnetic field (<500 G) directed along the z axis, OB can also be seen in MBBA at room temperature at a low power ($I_{th}<500$ W/cm^2).

Since the first observation made by Fréedericksz in 1927, all the observed Fréedericksz transitions (molecular reorientations) in NLC's are second order. This study showed that the first-order molecular reorientation and optical bistability can always be enhanced and hence always be seen in all nematics. The as-yet-unobserved first-order transition should be a viable candidate for the achievement of room-temperature, low-power, and externally controllable optical switches.

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[5]See Ref. 2 for a detailed discussion on the variational procedure for the case of $\mathbf{H}=0$.

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