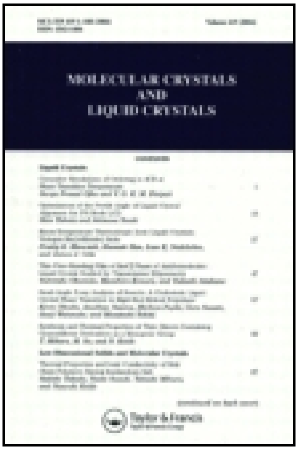

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Nonlocal Transverse Dependence of Molecular Reorientation Induced in a Nematic Liquid Crystal by a Guassian Laser Beam

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We present an analytical treatment of the nonlocal transverse dependence of the molecular reorientation induced by a Gaussian laser beam in a nematic liquid crystal. In the limit of small laser beam width compared to the film thickness, the induced molecular reorientation's transverse dependence exhibits broadening effect. These results are significant in opto-optical switching and self-phase modulation bistability.

INTRODUCTION

Optical and dc-plus-optical field induced molecular reorientation in nematic liquid crystal has recently received considerable attention.[1-3] Fundamental studies on the details of the process, and applied studies on optical wave front conjunction,[4] bistability[5]...etc. have been carried out. In almost all the optical field induced reorientation calculations and experimental studies, laser beams of finite beam waists are employed. Since the molecular reorientation effect in nematics is a nonlocal effect (i.s. the reorientation at a point in the liquid crystal depends on the values at all other points), the response of the liquid crystal to the optical field will assume in general a different spatial variation from the laser intensity profile. Since the molecular torque that counters the applied optical field torque depends on the spatial derivative of the reorientation angle, it is obvious that the transverse spatial response of the medium will be significantly affected by the boundary effect when the transverse dimension of the laser beam is comparable or smaller than the film thickness. Although there have been several attempts[6] at accounting for the finite beam size of the laser, they invariably make an over simplified assumption where the laser transverse intensity profile is taken as a rect function (i.e. finite within a radius ≈ beam waist (w_o) and zero outside). While this kind of assumption could help elucidate some effects associated with the beam size, it will not answer question on effects that depend on the *spatial variation* of the beam intensity. Self phase modulation effects, which result in spatial rings and transverse intensity distribution bistability, require a more exact description of the transverse spatial dependence.[7]

In this paper, we present an explicit calculation of the molecular reorientation's transverse dependence for two frequently employed geometries of interaction between the laser and the nematic liquid crystal film, as depicted in figures 1a and 1b. The angle β is non-vanishing. This geometry is required if the laser intensity is below the optical Freedericks threshold, in order to create director axis

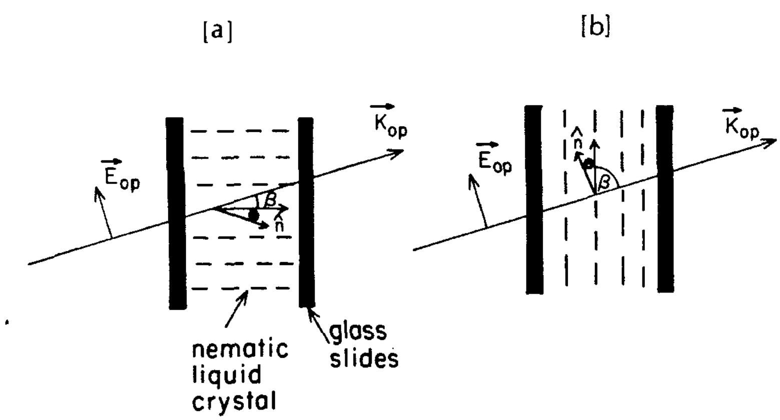


FIGURE 1 Schematic of the interaction geometry of a laser with propagation wave vector K_{op} and optical field polarization E_{op} with the nematic director axis n. (a) a homeotropically aligned sample (b) a planar sample.

reorientation. We will assume that β is nevertheless small enough that the problem is cylindrically symmetric along z. The radial distance from the axis z is designated as r. Specifically, we seek to compare and contrast the transverse spatial (r-dependent) profile of the induced reorientation with an applied transversely Gaussian laser beam.

Equation of torque balance and solution

Following the usual treatment of nematic reorientation, the free energy density of the system[8] is given by

$$F = \frac{1}{2} K_1 [\nabla \cdot \vec{n}(r)]^2 + \frac{1}{2} K_2 [\vec{n} \cdot \nabla \times n(r)]^2$$

$$+ \frac{1}{2} K_3 [\vec{n} \times \nabla \times \vec{n}(r)]^2 - \frac{\Delta\epsilon}{8\pi} [E \cdot \vec{n}(r)]^2 \quad (1)$$

where K_1, K_2 and K_3 are the elastic constants, $\Delta\epsilon$ the optical dielectric anisotropy, Eop the amplitude of the applied optical field and $\vec{n}$ is the director axis unit vector.

In the one-constant approximation ($K_1 = K_2 = K_3$), the elastic terms in F becomes[8] $1/2\ [K (\vec{\nabla} \cdot \hat{n})^2 + (\vec{\nabla} \times \hat{n})^2]$. The optical energy term gives $-\frac{\Delta\epsilon}{8\pi} E_{op}{}^2 \sin^2(\theta + \beta)$ (c.f. Figure 1). Minimization of the free energy with respect to variation in θ using a standard variational technique gives:

$$K \nabla^2 \theta + \frac{\Delta\epsilon}{4\pi} E_{op}{}^2 \sin(\theta + \beta) \cos(\theta + \beta) = 0 \quad (2)$$

Rewriting in terms of E_F [$E_F{}^2 = 4\pi^3 K \Delta\epsilon^{-1} d^{-2}$], the Fredericks field, one gets:

$$\nabla^2\theta + \alpha\, e^{-ar2}\, \theta + pe^{-ar2} = 0. \quad (3)$$

where

$$\alpha = \frac{\pi^2}{d^2} \cos 2\beta \frac{E^2}{E_F{}^2}$$

$$p = \frac{\pi^2}{2} \frac{\sin 2\beta}{d^2} \frac{E^2}{E_F{}^2}$$

and $E_{op}(r) = E\ e^{-ar/2^2}$ [with $a = 2/w_o^2$] is the incident gaussian laser beam's electric field of beam waist w_o.

Since the incident laser beam propagation direction deviates from being normal to the film (i.e. $\beta \neq 0$), the laser beam's transverse profile is, strictly speaking, non-cylindrically symmetric. Without much loss of accuracy in the final outcome of the physics of this problem, we shall nonetheless use a cylindrical-symmetric representation for both E(r) and ∇^2. The error made amounts to replacing a $\cos^2\beta$ factor by 1, which is not significant for small β. On the other hand, the effects (as measured by the width of the response $\theta(r)$ compared to the width of the incident laser beam) is rather substantial (>1). Equation (3) then becomes:

$$\frac{d^2\theta}{dr^2} + \frac{1}{r}\frac{d\theta}{dr} + \frac{d^2\theta}{dz^2} + \alpha\, e^{-ar^2}\,\theta + pe^{-ar^2} = 0 \tag{4}$$

Using the hard boundary conditions ($\theta = 0$ at $z = 0$ and $z = d$), the reorientation angle θ in the small θ limit can be approximated by the form:

$$\theta = \theta(r) \sin\frac{\pi z}{d} \tag{5}$$

substituting (5) into (4) and integrating with respect to z yields an equation in $\theta(r)$

$$\frac{d^2\theta(r)}{dr^2} + \frac{1}{r}\frac{d\theta(r)}{dr} + \left(\alpha e^{-ar^2} - \frac{\pi^2}{d^2}\right)\theta(r) + \frac{\pi}{2}\, pe^{-ar^2} = 0 \tag{6}$$

From known results in previous calculation, if $E << E_F$, then $\alpha << \pi^2/d^2$, and also $\alpha << \beta$ (which implies also $\alpha\,\theta << \beta$).
Then equation (6) reduces to a more manageable form

$$\frac{d^2\theta(r)}{dr^2} + \frac{1}{r}\frac{d\theta}{dr} - \frac{\pi^2}{d^2}\theta(r) = -\frac{\pi p}{2} e^{-ar^2} \tag{7}$$

The boundary condition for $\theta(r)$ are

$$\theta(r) = \text{finite at } r = 0$$

$$\theta(r) \rightarrow 0 \text{ as } r \rightarrow \infty \tag{7a}$$

It is important to point out here that we have taken into account the actual transverse dependence of the optical field E(r). This is different from the treatment of Csillag et al., who used a rect function E = constant for $r < w_o$; $E = 0$ for $r > w_o$. Furthermore, we are considering the case where $\beta \neq 0$, and $E << E_F$, while Csillag et al.'s calculation is based on the special *case* $\beta = 0$, where, in order to get non-vanishing θ, *E must be greater than* E_F.

The solution for $\theta(r)$ as described in equation (7), subject to the boundary conditions (7a) can be obtained using a straight forward Green's function technique.

$$\theta(r) = \frac{\pi p}{2}\left[Ko\left(\frac{\pi r}{d}\right) \int_o^r xe^{-ax^2} I_o\left(\frac{\pi x}{d}\right) dx + I_o\left(\frac{\pi r}{d}\right) \int_r^\infty xe^{-ax^2} K_o\left(\frac{\pi x}{d}\right) dx \right] \tag{8}$$

where $I_o(y)$ and $K_o(y)$ are the Hyperbolic Bessel Functions.[9] The asymptotic behaviour of these two functions are:

$$r \to o,\ I_o(y) \to 1,\ Ko(y) \to -\ln y;$$
$$r \to \infty,\ I_o(y) \to \frac{e^y}{\sqrt{y}},\ K_o(y) \to \frac{e^{-y}}{y}. \tag{9}$$

Note that the solution of $\theta(r)$ given in (8) obeys the requirement that $\theta(r)$ is finite at $r = 0$ and $\theta(r) \to 0$ for larger r.

DISCUSSION

The physical effect associated with the boundary effect in r is particularly transparent when we evaluate $\theta(r)$ for large values of a, i.e. small beam waist compared to the film thickness d. For convenience, we choose $d = \pi$ unit and write the two integrals in the curly bracket in (8) as J_1 and J_2, respectively. In general, the integrals have to be numerically evaluated. However, in the case of large a, one can analytically approximate (8) by asymptotic expansions and gain some interesting insights.

For example, consider $a = 900$. In that case one notes that the exponential factor e^{-ax^2} in the integrands will drop to vanishing value for value of $x > \frac{1}{\sqrt{a}}$, i.e. for $x > 0.03$. On the other hand, as long as we are interested in $\theta(r)$ for $r < 0.3$ (which is >> 0.03), we can use the asymptotic forms of Io and Ko for small argument (c.f. eqn. 9), we get

$$\theta(r) \sim \frac{\pi p}{2}\left[-\ell n\, r \int_0^r x e^{-ax^2}\, dx + \int_r^R x e^{-ax^2} (-\ell n\, x)\, dx\right]$$

$$= \frac{\pi p}{2}[J_1 + J_2] \tag{10}$$

where the upper limit R large in J_2 is the value of r large enough for e^{-ar^2} to go to vanishing value, yet small enough for the asymptotic form [$-\ln x$ for Ko(x)] to be valid, as mentioned earlier. Without any loss of accuracy we will henceforth set the factor $\pi p/2$ to be unity in the following discussion.

Figures 2a and 2b is a plot of the two integrals J_1 and J_2 in (10), for $a = 900$. Notice that J_2 as a function resembles much the input Gaussian beam profile, with a half-width close to the halfwidth of the input laser beam width $\left(\frac{1}{\sqrt{a}}\right)$. On the other hand, the function J_1 is vanishing at $r = 0$, peaks at near $r = \frac{1}{\sqrt{a}}$, and "decays" monotonically and "slowly" at large r. This function depicts the effects of the boundary value imposed at large r, and the molecular torque from molecules situated at distances outside the laser beam radius. The net results of these two vastly different functions is a somewhat "Gaussian" in form function for $\theta(r)$, plotted in dash line. The solution for $\theta(r)$ clearly shows that it has a half width of several times $\frac{1}{\sqrt{a}}$. In fact the function drops to e^{-1} of its central maximum (at $r = 0$) at a radial distance of $r_o = 0.25$, which is about 7 times the laser beam width.

Similar broadened radial dependences for $\theta(r)$ are obtained for large values of a (when the above integrals based on the asymptotic approximations for I_o and K_o are valid). For $a \approx 100$, for example, we get a "broadened" $\theta(r)$ with a e^{-1} width of 0.46 which is about 4.6 times the incident laser beam width.

We have quantitatively numerically (i.e. not using any asymptotic approximations) evaluated the R.H.S. of equation (8) for values of

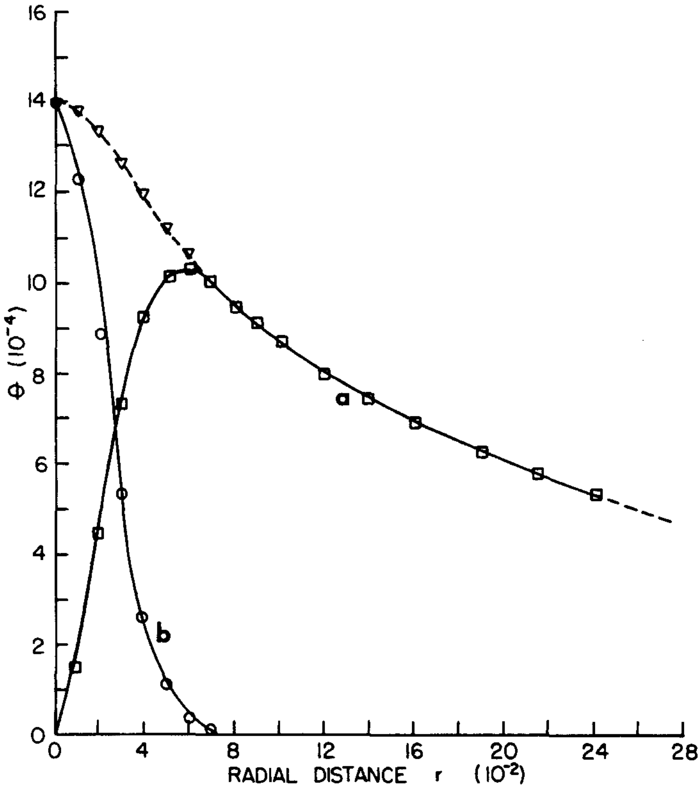


FIGURE 2 Plot of the two functions J_1 and J_2 comprising $\theta(r)$. The parameter $\pi p/2$ is set equal to unity.

a ranging from 0.15 to 500. Results for a ≈ 100 and greater agrees very closely with the above analytical deductions. Figure 3a (dotted line) shows the numerical results for a ≈ 100 using (8). The e^{-1} value of $\theta(r)$ occurs at a radial distance of r ≈ 0.55. Also plotted on figure 3 in full line is a Gaussian function $\exp \frac{-r^2}{0.55^2}$ for comparison purpose. It is interesting to note that from r = o to r = 0.55, $\theta(r)$ follows quite closely the Gaussian form, but drops off much slower than a Gaussian for radial distances greater than the new half width. In general, for smaller value of a the ratio of the computed half width to the input laser width decreases.

Figure 3b and 3c for example show the numerical plot for a = 1 and a = 0.5, respectively. The e^{-1} value for $\theta(r)$ occurs at r very close to 1 ($r_o \approx 1.6$ for both cases). These results are summarized in

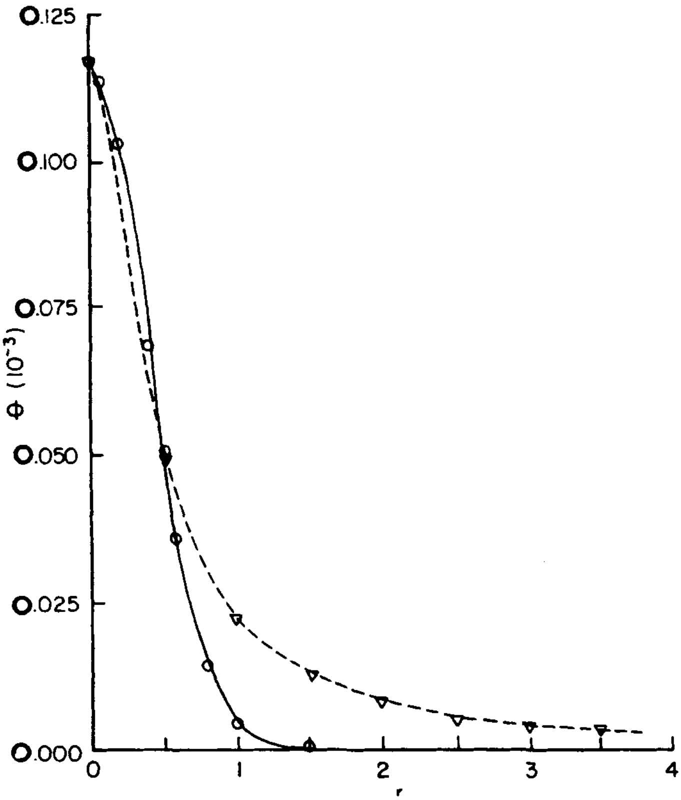
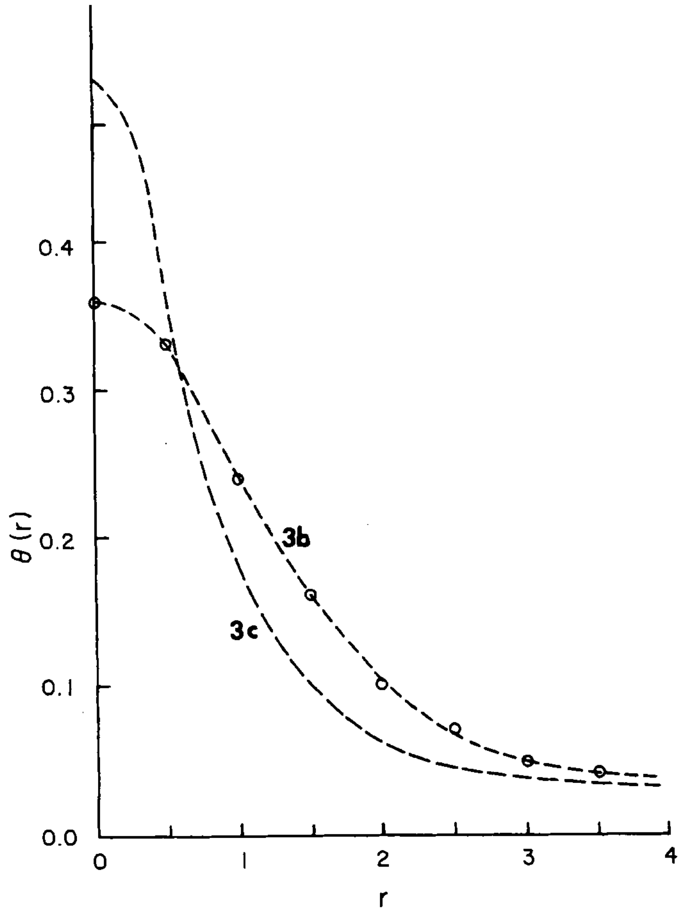


FIGURE 3a Dash-line: numerical results of $\theta(r)$ based on equation (8) for a = 100, for small β and for $E_{op} << E_f$. The plot shows a width of about 0.55. For comparison, a Gaussian function $\exp - r^2/(0.55)^2$ is also plotted. 3b. Plot of $\theta(r)$ for a = 1 for $\beta << 1$ and $E_{op} << E_f$. 3c. Plot of $\theta(r)$ for a = 0.5 for $\beta << 1$ and $E_{op} << E_f$.

figure 4, which shows a logarithmic plot of the "normalized" width of $\theta(r)$ function $\left(\text{with respect to the width to the input width } \frac{1}{\sqrt{a}}\right)$ for values of a ranging from very small (~ 0.15) to very large (900). For small value of a, the width of θ approaches the input laser beam width.

Another point to note is the maximum response of the system to the optical field, i.e. $\theta(o)$. From equation (6), one can see that the torque due to the surrounding (i.e. around the laser beam) molecule becomes insignificant compared to the torque form the boundary plates when a is small (i.e. laser width is large compared to d



when $\frac{d^2\theta}{dr^2}$ and $\frac{1}{r}\frac{d\theta}{dr}$ are both $<<(\pi^2/d^2)\theta)$. In that case, $\theta(o) \approx \frac{\pi p}{\pi^2}\left(\frac{d^2}{2}\right) = \frac{\pi p}{2}$ since we set $d = \pi$. On the other hand, when a is large, the molecules surrounding the beam waist has a much greater total effect on the molecular reorientation. $\theta(o)$ can be deduced from (10) to be given by $|\theta(o)| \approx \frac{\pi\beta}{2}\left(\frac{1}{a}\right)$. This is smaller than the $\theta(0)$ for small a limit by a factor of about $\frac{1}{ad^2} = \frac{w_o^2}{d^2}$. The numerical values of

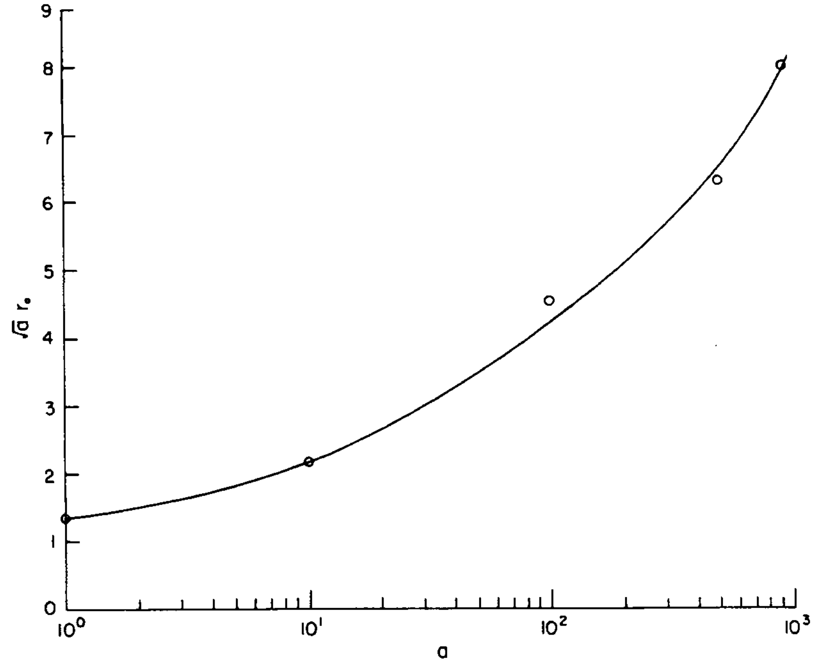


FIGURE 4 Plot of the ratio of the e^{-1} width (r_o) of $\theta(r)$ to that of the input laser ($1/\sqrt{a}$), versus the log of the numerical value of a.

$\theta(o)$ confirm this result. For $a = 900$, $\theta(o) \sim 10^{-3}$; for $a = 100$, $\theta(o) \sim 10^{-2}$ and for $a = 1$, $\theta(o) \sim 1$, following roughly the a dependence.

Finally, we would like to determine under what conditions are the theoretical effects obtained here experimentally observable. The best experimental observational method is the far field diffraction rings associated with the transverse phase shift, as studied by Durbin et al. The magnitude of the phase shift that is produced by the Gaussian beam is thus a good indicator of whether the predicted effect is observable. To get some real numbers, we note here that in typical experiments where the orientational nonlinearity of liquid crystal is observed, $\beta = 0.3$. If we use $E_{op} \sim 0.3\ E_F$, we get $p \sim (0.3)\ (0.3) \sim 0.1$. For $a = 1$, $J_1(o) + J_2(o) \sim 1$. Hence $\theta(o) \sim \pi p\ (J_1 + J_2) \sim 0.1$.

This reorientation gives rise to a change in the refractive index $\delta n \sim \Delta\epsilon\ 2\ \beta\theta \sim 0.04$. We have let $d = \pi$ units in the calculation. If this corresponds to a thickness of 100 μm, then the phase shift associated with δn is $\frac{2\pi}{\lambda}\, d\ n \sim 8\ \pi$.

For a = 10, J_1 (o) + J_2 (o) ~ 10^{-1}, θ(o) ~ 10^{-2}, and the phase shift is 0.8π. Since for values of a ranging from 1 to 10, the phase shift is large (and easily observable), while the broadening effects (c.f. figure 4) for this range of a are also substantial, experimental observation of the predicted broadening effects is best done under these geometrical conditions.

There is also a practical reason not to pursue the large a limit in practice, besides the fact that larger a means smaller θ. For a = 400, for example, this means $w_o^2 = \frac{2}{400}$ i.e. $w_o \cong 0.07$ unit. Since $d \doteq \pi$ unit corresponding to 100 μm, say, we have $w_o = \frac{(0.07)}{\pi} \times 100\ \mu m = 2.23\ \mu m$. If a laser is focused to this spot size, the confocal parameter $Z_o = \frac{\pi w_o^2}{\lambda}$, which roughly measure the distance in which the beam is parallel, will be on the order of $\frac{\pi(2.23\ \mu m)^2}{0.5\ \mu m} \sim 14\ \mu m$. That is, the beam is no longer parallel within the film. For a = 10, however, the same calculation gives a confocal parameter $Z_o \cong 560\ \mu m$, which clearly means the beam is parallel (i.e. has the same size) throughout the thickness of the film.

REMARKS

In conclusion, we have quantitatively evaluated the effect of the nonlocal response of the nematic film to an input Gaussian laser beam. The torque due to molecules at large radial distances from the laser beam gives rise to, in general, a broadened radial function for the reorientation. Although this may be expected from a general consideration, the explicit quantitative details obtained here allow one to calculate accurately the transverse phase shifts. This is important, for example, in opto-optical modulation, where one laser beam is used to induce a refractive index change, and therefore a transverse phase shift, on another laser beam and the far field transmitted beam intensity exhibit spatial redistribution or switching. Another area where this will be important is in the recently demonstrated transverse phase shift induced optical bistability of a single laser beam,[5] where the switching phenomenon depends very sensitively on the laser beam waist and the width of the transverse phase shift induced by the laser. The results of the present calculation is presently being incorporated in the theory and experiments of these two effects, and will be reported later.

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