# Reorientation with a micron lattice constant in unsteady optical four-wave mixing in a nematic liquid crystal

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A four-wave mixing of a ruby laser beam ($\tau \sim 10^{-3}$ s) is used to excite and then to detect reorientation space lattices of a nematic liquid crystal with an extremely small lattice constant $\Lambda$, reaching values as small as $\Lambda/2\pi = 2.7\times10^{-5}$ cm.

Orientational optical nonlinearity of nematic liquid crystals,[1–4] which has recently attracted increasing attention because of the large values of the effective cubic susceptibility, has nonetheless a serious drawback in terms of applied use: The response of reorientation $\theta$ of the director of a nematic liquid crystal to an electric pump field of a light wave is nonlocalizable. This nonlocalizable response stems from a quadratic dependence of the elastic energy of spatially nonuniform reorientation on the wave number $q$ of the reorientation, which sharply decreases the steady-state amplitude of the reorientation upon an increase in $q$. A nonlocalizable response of this sort occurs, however, only in the case of a steady deformation. If, on the other hand, the time required for the nematic liquid crystal to respond to light is $t \ll \tau_q$, where $\tau_q$ is the time it takes the amplitude of the reorientation for a given $q$ to reach a steady state, then the reorientation is determined solely by the orientational viscosity, rather than by the elastic energy, and is thus independent of $q$. In the present letter we study the orientational nonlinearity of a local nature, caused by an unsteady reorientation of this sort, in the particular case of a four-wave mixing of waves of various types of polarization (Ref. 1) in a nematic liquid crystal.

We assume that oppositely directed plane waves $\mathbf{E}_1 = \mathbf{e}_y E_1(t)\exp(i\mathbf{k}_e\mathbf{r} - i\omega t)$ and $\mathbf{E}_2 = \mathbf{e}_y E_2(t)\exp(-i\mathbf{k}_e\mathbf{r} - i\omega t + i\varphi(t))$, [where $\mathbf{k}_e = (2\pi n_\parallel/\lambda)\times(\mathbf{e}_x\alpha + \mathbf{e}_z)n_{\parallel,\perp}$ are the refractive indices of light which is linearly polarized parallel to or perpendicular to the director, respectively; $\lambda$ is the wavelength of light in a vacuum; and $\alpha$ is a small angle] and a weak signal wave $\mathbf{E}_3 = \mathbf{e}_x E_3(t)\exp(i\mathbf{k}_0\mathbf{r} - i\omega t)$, where $\mathbf{k}_0 = (2\pi n_\perp/\lambda)\mathbf{e}_z$, propagate in a planar sample of a nematic liquid crystal with an unperturbed director $\mathbf{n}^0 = \mathbf{e}_y$ (where $\mathbf{e}_i$ is the basis vector in a Cartesian system). The $z$ axis is perpendicular to the walls of the sample ($0 \leqslant z \leqslant L$); an increment in the phase, $\varphi(t)$, is introduced in order to demonstrate that there is no time-dependent coherence of wave $\mathbf{E}_2$ with the waves $\mathbf{E}_1$ and $\mathbf{E}_3$. In such a geometry the interference of waves $\mathbf{E}_1$ and $\mathbf{E}_3$ leads in the nematic to an excitation of a spatial-periodic reorientation of the director $\delta\mathbf{n} = \theta\mathbf{e}_x$ (see Ref. 1) with a wave vector $\mathbf{q} = \mathbf{k}_e - \mathbf{k}_0$. The scattering of $\mathbf{E}_2$ by the lattice of the dielectric tensor $\delta\epsilon_{ik}$ produced by this reorientation generates a wave $\mathbf{E}_4 \propto \mathbf{E}_1\mathbf{E}_2\mathbf{E}_3^*$ which is phase conjugated with the wave $\mathbf{E}_3$.

The dynamic equation for the reorientation of the director in this geometry is[1]

$$\eta\frac{\partial\theta_m}{\partial t} - (K_{11}\alpha^2 n_\parallel^2 + K_{22}(n_\parallel - n_\perp)^2)\frac{4\pi^2}{\lambda^2}\theta_m = \frac{\epsilon_a}{16\pi}E_1E_3^* . \tag{1}$$

Here $\eta$ is the orientational viscosity of the nematic, $K_{ii}$ are the Frank constants, and $\epsilon_a = n_\parallel^2 - n_\perp^2$; the solution is sought in the form $\theta = \theta_m(t)\exp(i\mathbf{q}\mathbf{r})$. In the transient regime, in which all changes occur in a time $t \ll \tau_q \sim (\eta/K_{22}q^2)$, the elastic term in (1) can obviously be dropped. In its simplified form Eq. (1) can now be easily solved:

$$\eta\frac{\partial\theta_m}{\partial t} = \frac{\epsilon_a}{16\pi}E_1E_3^* , \tag{2}$$

$$\theta_m(t) = \frac{\epsilon_a}{16\pi\eta}\int_0^t E_1(t')E_3^*(t')dt' . \tag{3}$$

Equation (3) clearly shows that the reorientation amplitude is independent of $\alpha$ or, equivalently, that it is independent of $q$ for all $q$ that satisfy $\tau_q \gg t$; i.e., the dependence of $\theta_m$ on the exciting field is local.

The truncated wave equation for the wave $\mathbf{E}_4 = \mathbf{e}_x E_4(\mathbf{r},t)\exp(-i\mathbf{k}_0\mathbf{r} - i\omega t)$ can be written

$$\frac{\partial E_4}{\partial z} = -\frac{i\pi\theta_m(t)}{\lambda n_\perp}E_2(t)\exp(i\varphi) . \tag{4}$$

Using $\theta_m(t)$ from (3) and setting $E_4(x,y,L,t)=0$, we find

$$E_4(z=0;t) = -\frac{i\epsilon_a^2 L E_2(t)\exp(i\varphi)}{16\eta n_\perp \lambda}\int_0^t E_1(t')E_3^*(t')dt' .$$

Since in the actual experiment the waves $\mathbf{E}_{1,2,3}$ are formed by a single laser pulse $\sim 1$ ms in duration, we have, within unimportant differences in the terms, $E_2(t)=\sqrt{\xi_2}E_1(t)$ and $E_3(t)=\sqrt{\xi_3}E_1(t)$ in the Born approximation used by us. Switching to the instantaneous coefficient $R_I$ for the change in the intensity of the wave $\mathbf{E}_3$, we can therefore write

$$R_I(t) = \frac{|E_4(z=0,t)|^2}{|E_3(t)|^2} = \left[\frac{\epsilon_a^2 L}{16\eta n_\perp \lambda}\int_0^t E_1(t')E_2(t')dt'\right]^2 . \quad (5)$$

Before describing the experiment, we will make another important observation. Since $\theta_m$ does not depend on $q$, it is easy to see that when $\mathbf{E}_3$ is not a plane wave but has a finite angular spectrum, expression (5) remains in force, while the wave $\mathbf{E}_4$ is inverted exactly with respect to $\mathbf{E}_3$.

In the experiment we used a planar cell with a nematic liquid crystal 5 TsB of thickness $L = 140$ $\mu$m. The light source is a single-mode (in the transverse index) ruby laser ($\lambda = 0.6943$ $\mu$m) working in the regime of free-running pulses (the pulse length is $\tau \sim 1$ ms), with a pulse energy of $\sim 150$ mJ and a beam divergence $\theta_0 = \text{FWHM} \approx 6\times 10^{-4}$ rad. The pulse length is such that the transient condition $\tau < \tau_q$ for the reorientation lattices with a lattice constant $\Lambda \geqslant 1.4$ $\mu$m is satisfied, which corresponds to a convergence angle $\alpha_0$ of the $E_1$ and $E_3$ beams (in air) $\alpha_0 \lesssim 0.5$ rad. The laser beam is divided by polarization devices into an $e$-polarized beam $\mathbf{E}_1$ and an $o$-polarized beam $E_3$, whose power is one-tenth of the power of the $\mathbf{E}_1$ beam. These two beams cross in the sample which is placed at the focal point of the telescope consisting of lenses with a focal length $f = 25$ cm. The telescope is used to increase the power density of the reference waves $\mathbf{E}_{1,2}$ in the sample by compressing them to a diameter $a = \text{FWHM} \approx 150$ $\mu$m. The second reference wave is formed by reflecting the wave $\mathbf{E}_1$, which is transmitted through the telescope, from a mirror. The ratio of the intensities of the reference waves is $\xi_2 \doteq 0.5$. We measured the following parameters: $|\mathbf{E}_{3,4}(t)|^2$, $\int_0^t |E_1|^2 dt'$ (with the help of an integrating chain), the total energy of the inverted wave $W_4$, and the angular divergence of the waves $\mathbf{E}_3$ and $\mathbf{E}_4$ (by means of photometric measurements of the beam photographs at the focal point of the lens $f_1 = 100$ cm with the use of a stepped reducer).

We found that as the pulse energy $W_L$ is increased, a signal $\mathbf{E}_4$, which depends on $W_L$ in a nonlinear manner, is produced (Fig. 1). The shapes of the temporal envelopes $|E_3(t)|^2$ [Fig. 1(a)] and $|E_4(t)|^2$ [Fig. 1(b)] differ markedly, their difference being characteristic of time-varying four-wave mixing. We have measured $R_I(t)$ as a function of the square of the instantaneous value of the pulse energy $Q^2(t) \sim (\int_0^t |E_1|^2 dt')^2$ for various convergence angles, $\alpha_0$, of the beams [Fig. 2(a)]. We see that, irrespective of the total energy of the pulse and the angle $\alpha_0$, all results lie on a single straight line, consistent with (5). We have obtained instantaneous values of $R_I \approx 30\%$. In the

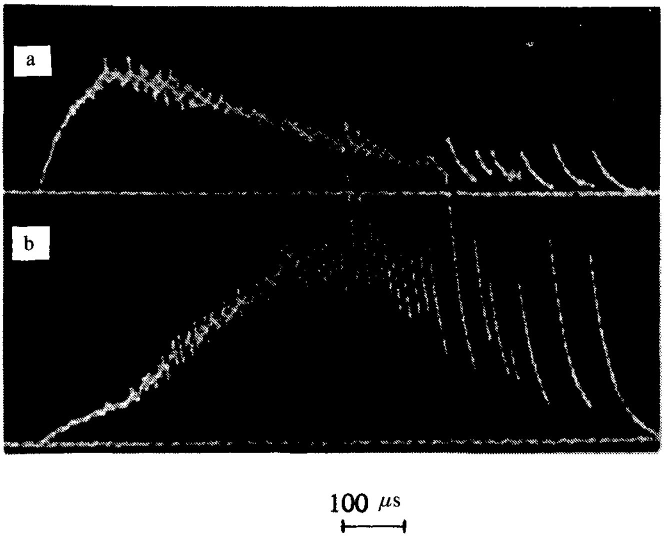


FIG. 1.

steady state $R_I$ for $\alpha_0 = 0.38$ rad should decrease by a factor of 25 in comparison with $R_I$ for $\alpha_0 = 0$.

The proportionality coefficient for this dependence is one-sixth of the theoretical coefficient calculated from (5) using a beam size $a = 150\,\mu$m. This discrepancy stems from the error in the overlap of the interacting beams, the error in the size $a$, and several other factors of a technical nature. The $W_L^2$ dependence of the energy reflection coefficient $R = W_4/W_3$ [Fig. 2(b)] is also linear, consistent with the theory if the shapes of the temporal envelopes for the various pulses are similar, an additional condition which is satisfied in our experiment. We have obtained values of $R \approx 10\%$.

The localizability of nonstationary orientational nonlinearity has made it possible to invert the wave front of the signal $\mathbf{E}_3$ with a finite angular spectrum. A phase plate,

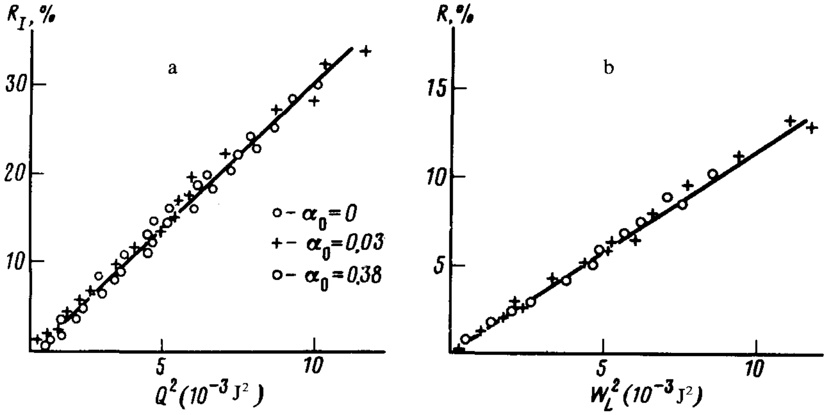


FIG. 2.

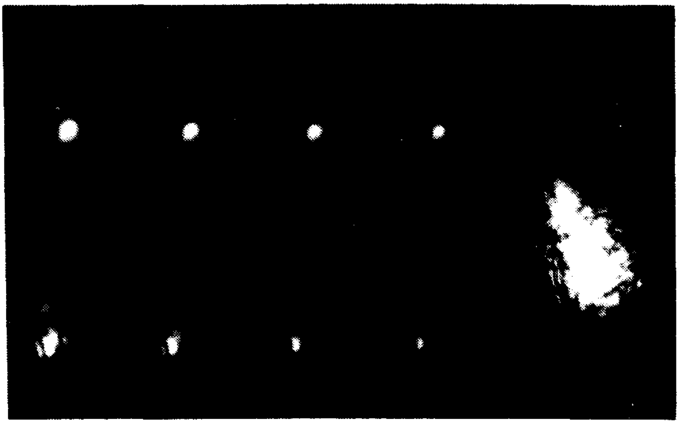

FIG. 3.

which degrades the initial beam divergence $\theta_0 = 6\times10^{-4}$ rad [Fig. 3(a)] to $3\times10^{-3}$ rad [Fig. 3(b)], is introduced into the beam $\mathbf{E}_3$. The energy reflection coefficient in this case remains constant, while the divergence of the signal $\mathbf{E}_4$, which is transmitted through the phase plate in the opposite direction, is corrected essentially to $\theta_0$, indicating that the wave front of the wave $\mathbf{E}_3$ has been inverted [Fig. 3(c)].

We have thus demonstrated that nonstationary orientational nonlinearity of nematic liquid crystals is highly localizable spatially. Because of this localizability, reorientation space lattices with a small (on the order of a micron) lattice constant can be effectively excited in these nematics. We have also used the nonlinearity to attain a wave-front inversion and a four-wave mixing.

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