Nonlinear dynamics in laser polarization conversion by stimulated scattering in nematic liquid crystal films

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We present a study of the nonlinear dynamics of a coherent polarization conversion process in nematic liquid crystal films. This effect is mediated by two-beam coupling between the incident polarized laser and its orthogonally polarized (lower-frequency) noise component scattered by the director axis fluctuations. A complete model for the director dynamics derived from the basic equations is presented. The existence of complex, time-, and intensity-dependent dynamics of the director motion such as oscillations and various bifurcations including period doubling is revealed.

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The study and modeling of nonlinear dynamics has become an important tool to gain understanding of optical solitons [1], self-organization [2], neural networks [3], pattern formation [4,5], and other complex physical systems. Liquid crystals [6,7], photorefractive materials [8], laser systems [9,10], and atomic ensembles [11] are just a few of the systems that exhibit and allow the study of nonlinear dynamic responses from a theoretical and experimental perspective. Liquid crystalline systems have become a preferred study ground for nonlinear effects because the collective dynamics of its director axis can bring forth an extremely nonlinear response, capable of photorefractive effects and grating formation [12,13], self-starting optical phase conjugation [14–16], polarization rotation and beam control, stimulated scatterings [17,18], and optically induced complex dynamics [19–23]. In turn, these effects allow the optical implementation of associative memories, image and signal processing [24], optically addressed spatial light modulators [25], and similar devices. Furthermore, liquid crystals also exhibit a large dielectric anisotropy and almost zero absorption from the visible to the near-infrared regime and beyond [6].

In this paper, we report theoretical studies of polarization conversion dynamics by two-wave mixing in a pure nonabsorptive nematic liquid crystal film [14,26], cf. Fig. 1 in which an extraordinary wave E_x is normally incident on a planar-aligned nematic liquid crystal cell. Scattering by the director axis fluctuations induces a lower-frequency orthogonally polarized component E_y inside the cell which will experience gain. The mixing of these two orthogonal waves creates a moving intensity grating responsible for the director axis reorientation (and refractive index grating). The driving field is strongly perturbed by the system because of energy coupling, and as the intensity of the light is increased, the director axis exhibits strong oscillations, as reported in recent experimental observations. A complete quantitative theory has been developed to account for severe pump depletion, and includes higher-order terms that have been neglected in previous steady-state treatments [26]. As shown in Fig. 2, the measurement of the temporal behavior of the output beams is straightforward and significantly easier than the four-detector polarimeter previously reported by Cipparrone *et al.* [27], or the study of optical nonlinearities through pattern formation such as reported in [4]. Potentially, the system will also provide a richer set of phenomena to study because it consists of two beams of slightly different frequency instead of one (compared to the homeotropic setup in [27]). The results are also reminiscent of instabilities present in optically injected semiconductor lasers, where nonlinear dynamics is bounded as a function of the injection level [9,10].

The equation that describes the evolution of the director $\hat{\mathbf{n}}=\cos\theta(z,t)\hat{i}+\sin\theta(z,t)\hat{j}$ can be deduced from the free energy of the system [12] consisting of a contribution from the elastic energy of the nematic and a term corresponding to the energy of the total electric field inside the cell $\mathbf{E}=E_x e^{i(k_x z-\omega_x t)}\hat{i}+E_y e^{i(k_y z-\omega_y t)}\hat{j}$. The small signal gain constant G will depend on the frequency difference $\Omega=\omega_x-\omega_y$, the elastic constant K_2, and the viscosity η, and is given by $G=f(\Omega)\pi\varepsilon_a^2/2cn_e\lambda K_2 q^2$, where $f(\Omega)$

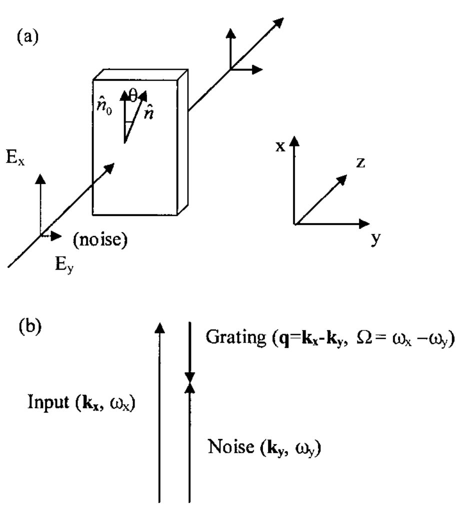

FIG. 1. (a) Depiction of a linearly polarized laser incident on a planar-aligned nematic liquid crystal. (b) Optical wave vectors k_x and k_y and grating wave-vector condition for maximum conversion.

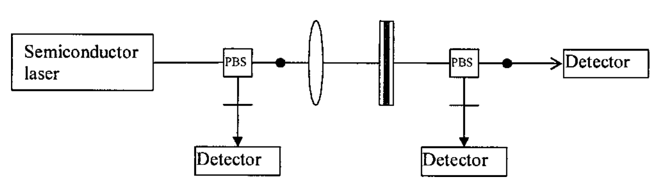

FIG. 2. Experimental setup. PBS, polarizing beam splitter.

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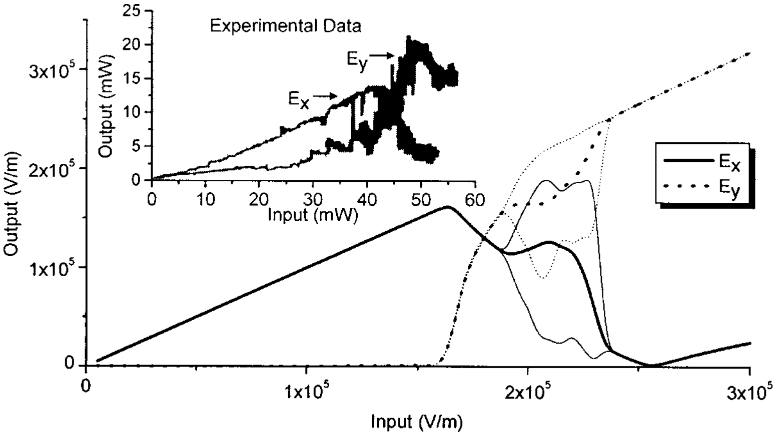

FIG. 3. Calculated output *e* and *o* waves as a function of the input field. Input laser is an *e* wave. Also shown is the experimental measured transmitted waves as a function of input laser power for a focused diameter of 40 μm.

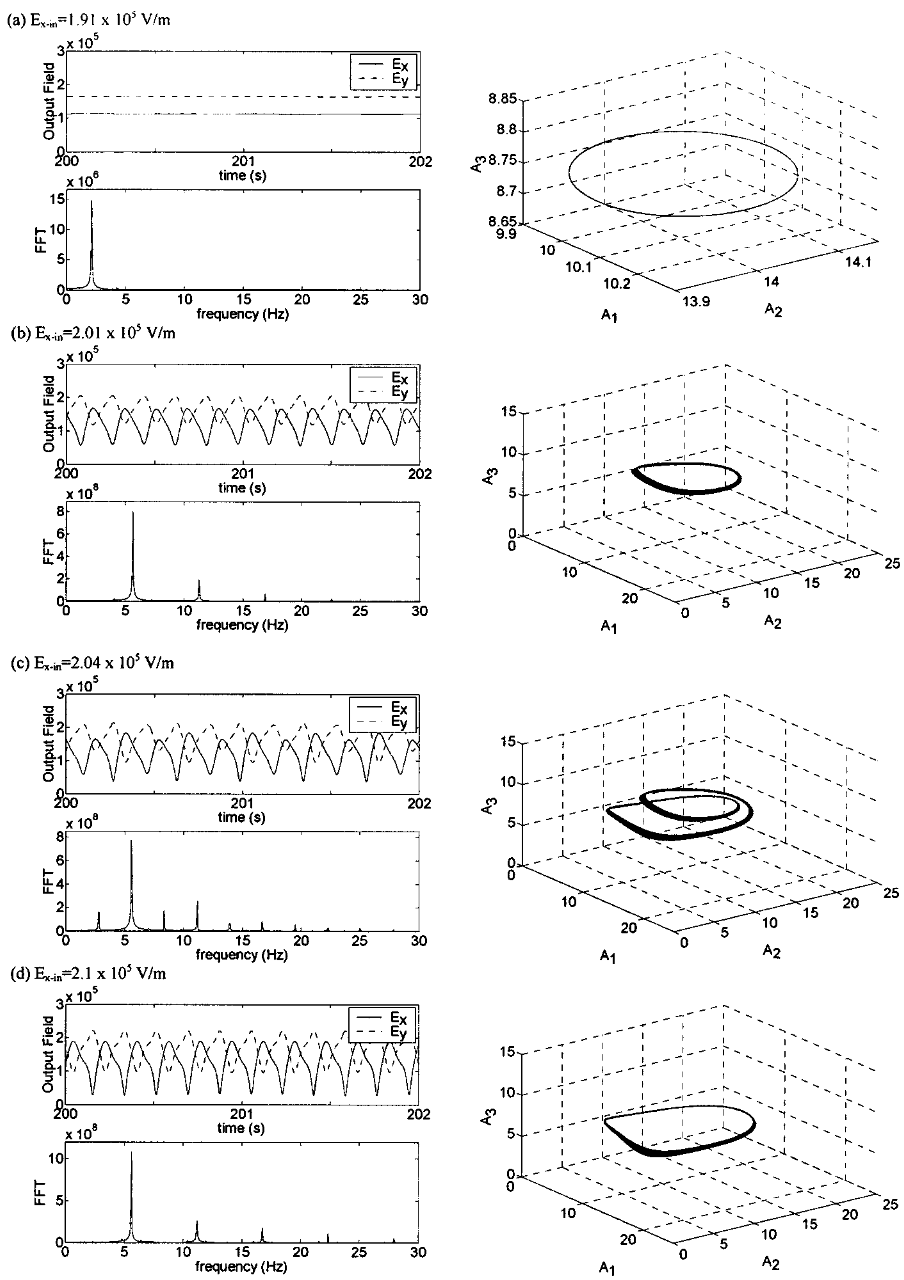

FIG. 4. (a)–(g) Output field dynamics, Fourier transform, and phase diagram of angle coefficients for different values of input *e*-wave field. Coefficients are given in mrads.

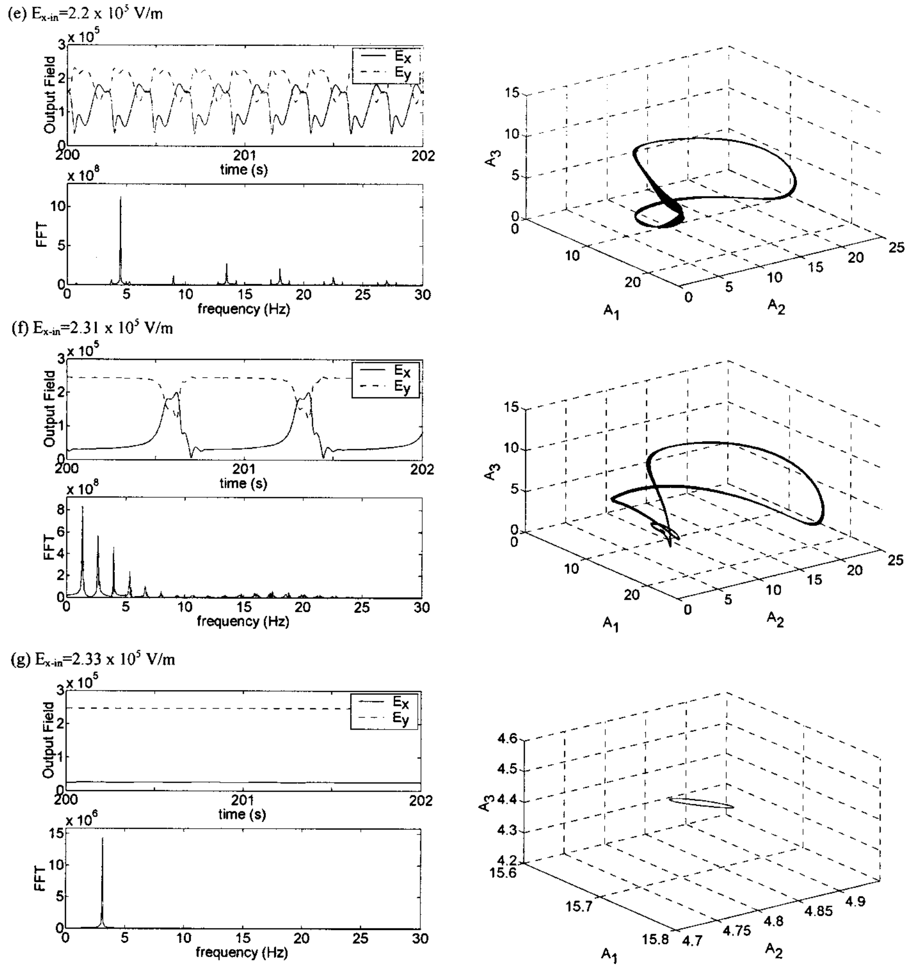

FIG. 4. (*Continued*).

$=(2\Omega/\Gamma)/(1+\Omega^2/\Gamma^2)$, $\Gamma=K_2q^2/\eta$, and q is the grating wave vector $\mathbf{q}=\mathbf{k}_x-\mathbf{k}_y$. Maximal gain occurs for $\Omega^2/\Gamma^2=1$.

Inserting the free energy into the Euler-Lagrange equation and using the dissipation function $R=\frac{1}{2}\eta\dot{\mathbf{n}}^2$ of the system with $\theta(z,t)$ as the independent variable, the equation of motion for the director angle can be shown to be

$$\begin{aligned}\eta(d\theta/dt)-K_2[\partial^2\theta(z,t)/\partial z^2]\\ =(\varepsilon_a/2)\,[\sin(2\theta)(|E_y|^2-|E_x|^2)\\ +\cos(2\theta)(E_xE_y^*e^{i(qz-\Omega t)}+E_x^*E_ye^{-i(qz-\Omega t)})].\end{aligned}\qquad(1)$$

The electric fields must furthermore satisfy Maxwell's equations. Writing $\theta(z,t)=\frac{1}{2}[\theta_s(z,t)e^{i(qz-\Omega t)}+\text{c.c.}]$ and using the slowly varying envelope approximation [i.e., $\partial^2\theta_s(z,t)/\partial z^2$ and $q\partial\theta_s(z,t)/\partial z$ are negligible compared to $q^2\theta_s(z,t)$], $\omega_y^2\gg\Omega\omega_y\gg\Omega^2$, and applying phase-matching conditions, the equations for the electric fields become

$$\partial E_x/dz=-i(\varepsilon_ak_x/4\varepsilon_x)|\theta_s|^2E_x+i(\mu\varepsilon_a\omega_y^2/4k_x)\theta_sE_y,\qquad(2)$$

$$\partial E_y/dz=i(\varepsilon_ak_y/4\varepsilon_y)|\theta_s|^2E_y+i(\mu\varepsilon_a\omega_x^2/4k_y)\theta_s^*E_x.\qquad(3)$$

In order to simplify the numerical analysis of the system, and under the assumption of strong anchoring, the director angle may be expanded as $\theta_s(z,t)\equiv\Sigma_nA_n(t)\sin(n\pi z/L)$. The director equation is then projected in each mode and integration is performed over the whole length of the cell, to obtain

$$\begin{aligned}\frac{dA_n}{dt}=\left\{i\Omega-\frac{K_2}{\eta}\left[q^2+\left(\frac{n\pi}{L}\right)^2\right]\right\}A_n+\frac{\varepsilon_a}{\eta L}\int_0^L\theta_s\sin\left(\frac{n\pi z}{L}\right)\\ \times(|E_y|^2-|E_x|^2)dz+\frac{\varepsilon_a}{\eta L}\int_0^L\sin\left(\frac{n\pi z}{L}\right)\\ \times\left(E_xE_y^*-\frac{1}{2}\theta_s^2E_x^*E_y-|\theta_s|^2E_xE_y^*\right)dz.\end{aligned}\qquad(4)$$

Only the three first terms of the expansion are considered since only the magnitudes of the first few modes are non-negligible, as the higher-order modes are strongly damped by elasticity [23].

The parameters used for the simulation correspond to the experimental results reported previously [26]. The liquid crystal used is E7 from EM Chemicals, with values $n_e=1.75$, $n_o=1.54$, elastic constant $K_2=3.0\times10^{-12}$ N, and viscosity coefficient $\eta=0.07$ P. For a wavelength of 1.55 μm (at which the absorption of E7 is negligibly small), the optimum gain coefficient corresponds to a frequency Ω

$=31$ Hz and defines a grating spacing of $1.55/(n_e-n_o)$ $=7.38$ μm. A 400-μm cell was considered.

The numerical computation of the dynamics of the wave mixing and director axis reorientation processes show a spectrum of effects. Only some exemplary ones are considered in this Brief Report. Figure 3 shows in thick lines the average value of the exit *e* and *o* fields as a function of the input field. The input *e* wave grows linearly at low input power while the *o* wave or "noise" component is nearly zero. After a threshold, the *o* wave will increase exponentially to a maximum value, and the *e* wave begins to show signs of depletion, eventually dropping to zero. After reaching the point of polarization conversion (at which the scattered *o* wave is greater than the exit pump beam), the system starts to oscillate, with the thin lines showing the minimum and maximum values the two fields can reach. This regime of self-oscillations ends when the input intensity is high enough so most of the energy in the input beam "switches" to its orthogonally polarized counterpart.

The oscillations observed just above the stimulated scattering threshold are reminiscent of director motion above the optically induced Fréedericksz threshold in previous studies. Accordingly, one would expect complex dynamical behavior in this regime. Indeed our calculations show a wealth of phenomena. Figure 4 highlights the exemplary results for different values of the field in the "oscillatory regime"—time evolution of the transmitted *e* and scattered *o* wave (E_x and E_y, respectively), the spectral components of the transmitted wave, and the dynamics in the A_n angle coefficient phase space. Three different cases may be considered: first, for intensities at the very start or end of the oscillatory regime [Figs. 4(a) and 4(g)] the oscillation is very small with a definite frequency (note that the phase and Fast Fourier transform diagrams for these two figures have a different scale from the rest). Second, Figs. 4(b) and 4(d) show a periodic regime with clearly defined oscillations for the output fields. Finally, and most interesting, Figs. 4(c), 4(e), and 4(f) show bifurcations and period-doubling occurring on the system. Also notice that the main frequency of oscillation is intensity-dependent. These results are in agreement with the oscillatory outputs (transmitted pump and signal beams) observed experimentally, cf. Fig. 3 inset. More recent experimental measurements also confirm the general behavior as described in Figs. 4(a)–4(g).

In conclusion, we have presented a complete theoretical treatment of the dynamics of multiwave mixing and director axis reorientation, and illustrated the possibilities of observing complex dynamical behavior in nematic liquid crystals driven by a coherent laser beam and its scattered noise. As shown in previous observations, the process of the orthogonally polarized noise building up to a coherent beam can be generated with an *o*-wave incident beam as well. It is also clear from the interaction dynamical equations that if the driving field or the nematic director axis is modulated, some "resonant" effects, multibifurcations, and chaotic responses could also arise. Effects such as various routes to chaos, control of the nonlinear dynamics, and the effect of different physical parameters in these processes are currently being studied both experimentally and theoretically.

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