

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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TRANSIENT AND STATIONARY WAVEMIXING AND INTERFACE SWITCHING WITH LIQUID CRYSTALS

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Abstract We present the results of recent theoretical and experimental studies of optical wave mixings and switchings based on the optical nonlinearities of nematic liquid crystals. These studies have demonstrated several new fundamental understandings of stationary and transient optical wave mixing and switching processes.

Introduction

Nonlinear optics of liquid crystals in their mesophases (cholesterics, smectics and nematics) have been vigorously studied in the past few years. Two detailed reviews of works done up to 1987 have appeared. The review by Tabiryan, Sukhov and Zeldovich¹ deals with the orientational optical nonlinearity, whereas Khoo's article² deals with several nonlinear processes associated with both the thermal and orientational nonlinearities of liquid crystals. The emphasis of this paper is slightly different, and is centered on studies of new aspects of two general nonlinear processes, where liquid crystals' extraordinarily large nonlinearities allow us to experimentally confirm these new theoretical observations. These new effects are generally observable in highly nonlinear optical thin films, and in liquid crystals in particular, and thus allow one to construct useful nonlinear optical devices, besides providing new insights into existing processes or devices.

The processes under study are (i) optical wave mixing of two or more coherent laser beams in a thin nonlinear film and, (ii) transmission/reflection switching in a dielectric cladded nonlinear thin film.

Optical Wave Mixing

Figure 1 depicts schematically the interaction of two coherent laser beams (E_1 and E_2) in a nonlinear thin film.

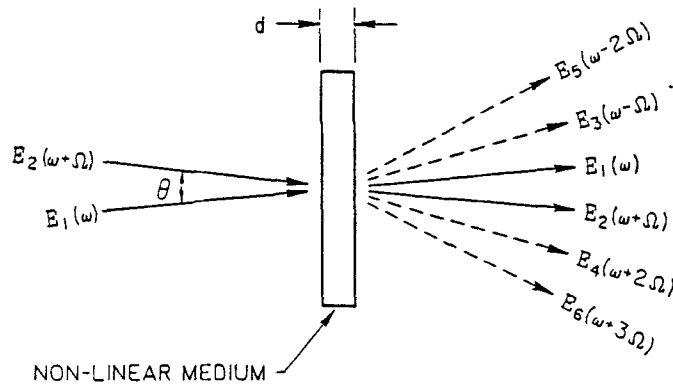


Fig.1

The interference sets up an intensity grating, which in turn generates an index grating via the nonlinear response of the medium. As a result of the index grating, several diffracted beams (E_3 , E_4 , E_5 , E_6) are produced at the exit side. Within the nonlinear medium, all these beams interact strongly with one another, generating new grating, phase shifts, energy exchanges,

etc. These intensity and phase coupling equations are of the general form.

$$\frac{\partial I_i}{\partial z} = g_i(I_i; I_j I_k I_l, \phi, \phi_j; t) \quad (1)$$

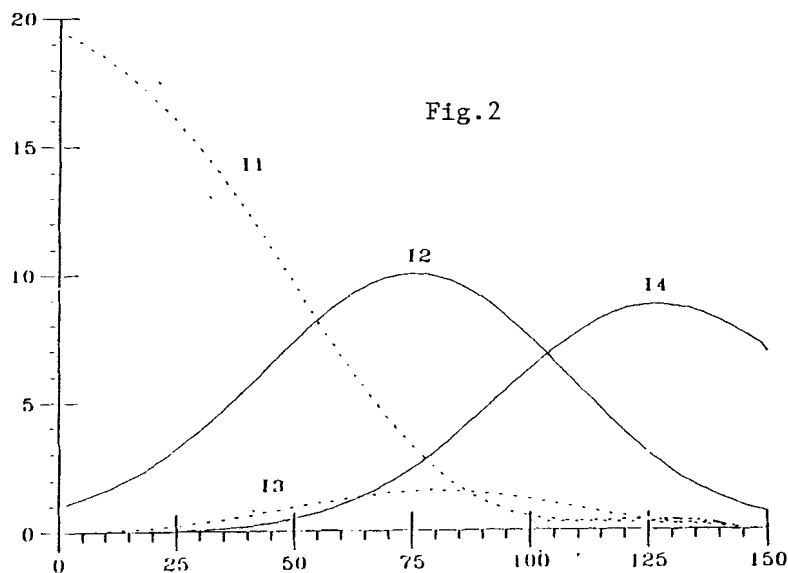
$$\frac{\partial \phi_i}{\partial z} = h_i(I_i, I_j I_k, \phi, \phi_j, t, \text{etc.}) \quad (2)$$

Among the many physical processes associated with these intensity and phase coupling effects is the amplification of the weak probe beam E_2 by the strong pump beam E_1 . Amplification of the probe beam can occur under the following conditions:

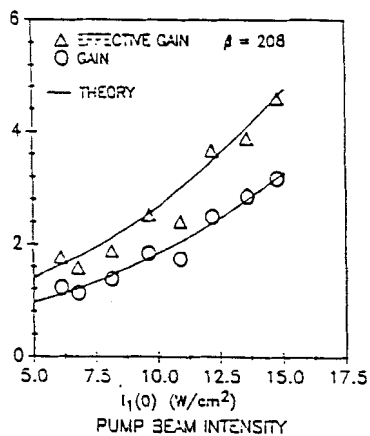
- (i) If the medium response is local, i.e., the refractive index grating coincides with the intensity grating, the amplification is via the scattering of the pump beam (from the pump-diffracted beam (E_3) grating) into the probe beam direction in the stationary case.
- (ii) If the pump and the probe beam frequencies are different, the cw probe beam will experience gain if it is stoke-shifted with respect to the cw pump. The frequency shift should be on the order of the inverse of the grating decay time.
- (iii) If the incident lasers are pulses with pulse duration shorter than the grating decay time, then transient phase shift between the intensity and the refractive index grating will give rise to amplification of the weak beam by the strong pump.

We have developed the theory and experiment corresponding to all these three cases. For cases (i) and (ii), the orientational nonlinearity of nematic liquid crystal is utilized. In particular, the result for case (ii) has been reported in reference 3 and a

detailed theory is given in reference 4. Figure 2 shows the dependence of the intensity of all the six beams as



a function of the distance z into the nonlinear medium, showing how the intensity exchanges, grows and decays, etc.⁴ In theory, self-phase modulation, mutual phase modulation, losses, phase mismatch, etc. are all accounted for. Using this theory, we obtain a very good fit with experimental results. Figure 3 shows the plots

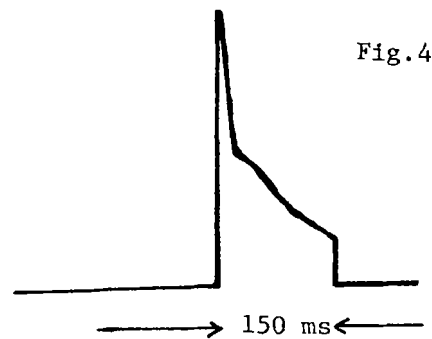


of the effective gain ($I_{\text{probe}}(\text{with pump})/I_{\text{probe}}(\text{without pump})$) and gain ($[I_{\text{probe}}(\text{with pump}) - I_{\text{probe}}(\text{incident})]/I_{\text{probe}}(\text{incident})$), and the solid lines are the corresponding theoretical curve. The experimental parameters used in this experiment are: laser used is 5145Å line of an Argon laser; liquid crystal sample is 120µm thick and homeotropically aligned; liquid crystal used is PCB (pentyl-cyano-biphenyl); laser polarization is 22° with respect to the director axis. A gain as high as 6 can be obtained.

Using the large thermal refractive index changes in liquid crystals, these amplification effects have also been observed with infrared (CO_2) lasers; probe gains as high as 20 can be obtained with laser intensity on the order of a few watts/cm² (reference 5).

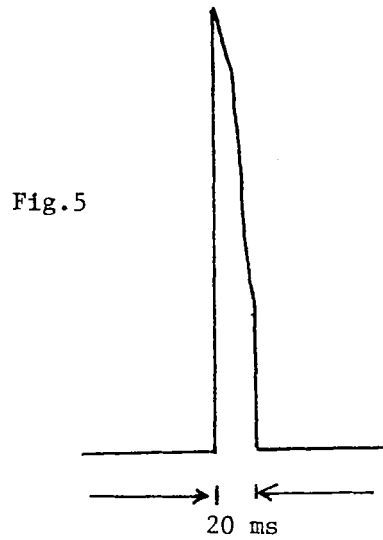
More recently, we have conducted detailed experimental studies of thermal grating mediated probe amplification with pulsed lasers, and observed effects associated with the transient phase shift. The experimental details are similar to those described in reference 5, except that square pulses are used. The pulse durations are varied from 10ms to 200ms. The crossing angle (in air) is 2.62°, corresponding a grating constant of 200 µm. The liquid crystal thickness is 125 µm. The thermal index gradient used is dn_o/dT . The grating decay time constant for this configuration is on the order of 7 milliseconds. This means one could expect transient 2-wave mixing effect (case (iii)) to manifest in this time scale.

Figure 4 shows a trace of the transmitted probe pulse for a square input pulse duration of 150ms. A well



defined spike is observed. The spike dies off completely in about 20ms, and is followed by a slower decaying tail. The initial amplification effect is due to the transient grating mediated wave mixing effect, whereas the slowly decaying tail is the four-wave mixing mediated probe amplification. The slow decay is the result of the thermal grating build up (owing to the laser induced rise in temperature and the increased dn_0/dT with higher temperature), which competes with the thermal diffusive

process. Figure 5 captures the initial transient



amplification of a 20ms square probe pulse. The rise and fall is in excellent agreement with the transient 2-wave mixing theory⁶ using a decay time constant τ of 7ms. We have conducted several other experimental and theoretical studies of these transient effects, such as angular dependence, intensity, intensity ratio, etc. Details will be published in a longer article elsewhere.

Interface switching

Figure 6 shows a schematic of optical switching at a dielectric (prisms) cladded nematic liquid crystal film.

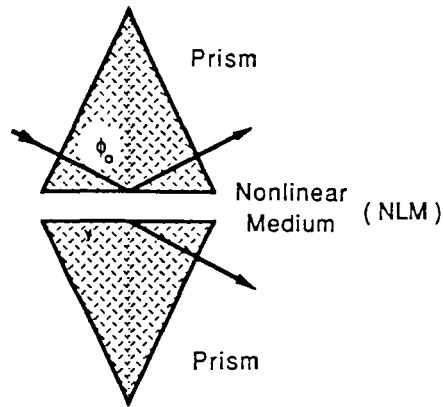


Fig.6

A laser is incident from the dielectric medium (glass prism) with refractive index n_0 at an angle ϕ_0 ($\phi_0 \approx \phi_{\text{TIR}}$, the angle for total internal reflection TIR). If the medium possesses a positive nonlinearity, it is obvious that switching from the TIR to the transmission state can occur if $\phi_0 \geq \phi_{\text{TIR}}$, as a result of the evanescent field induced positive refractive index change.

Case II

On the other hand, if the medium possesses negative nonlinearity, then switching from the transmission to the TIR state can occur if $\phi \leq \phi_{\text{TIR}}$, as a result of the laser induced negative refractive index change.

In case I, we have conducted a detailed theoretical study of the dynamics of the switching process. In our theory, both the medium's response time and the intense reflection feedback between the two interfaces are explicitly accounted for. The latter means that the thickness of the nonlinear film will matter considerably in the turn-on time, a feature that is ignored in the usual theory assuming infinite half-space. Using a modified (to include time-dependence) generalization of the transfer matrix developed in reference 7, this switching effect is studied with the following factors explicitly accounted for, namely, reflection feedback, angle of incidence, intensity, medium thickness, material response time (τ), refractive indices, etc. Figure 7 shows the time dependence of the transmission for four different incident angles ($\phi = 1.22, 1.23, 1.24$, and 1.25). As the curves clearly show, steady state transmissions are reached at times on the order of 0.1τ , 0.2τ , τ , and 4τ , respectively.

More results for both Cases I and II, and experimental observations with visible and infrared laser pulses will be the subject of longer articles elsewhere. This research is supported by an NSF grant ECS 871-2078.

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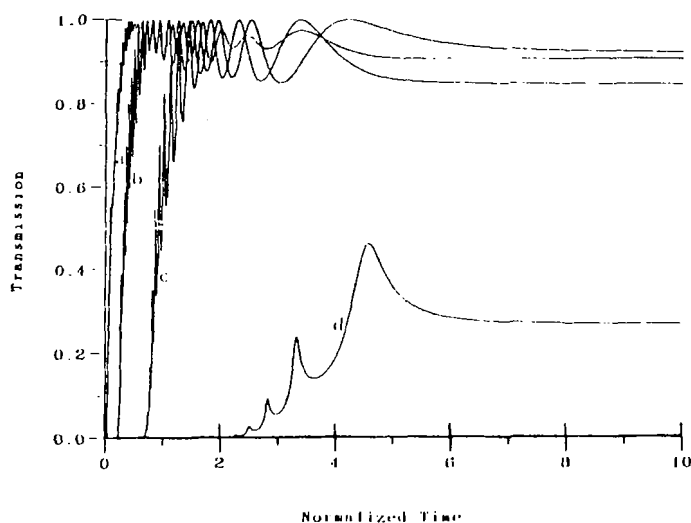


Fig. 7