
**PHOTOREFRACTIVITY AND NONLINEAR ELECTRO-OPTICAL
WAVE MIXINGS IN LIQUID CRYSTALS**

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Final Report

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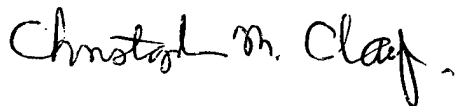
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13. ABSTRACT (Maximum 200 words) This research program was aimed at gaining a complete fundamental understanding of recently discovered nonlinear electro-optical effects in liquid crystalline systems, and exploring their applications in holographic storage, optical wave-mixing and phase conjugation beam/image processing and modulation devices. In particular, the mechanisms and dynamics of photo-charge generation, space charge fields and the resultant refractive index change and holographic grating formation in dye- and fullerene doped-nematic liquid crystal have been quantitatively investigated. This work has established such liquid crystalline materials as among the most sensitive nonlinear optical materials for performing holographic storage, wave-mixing, beam and imaging processing, and switching processes. Basic understanding of the photophysics, electro-dynamics and electro-optical properties of these liquid crystalline material systems will be important for related fields such as electro-optical flat panelled displays and imaging processing, and new material development. Investigation was made of nonlinear fiber arrays made by filling capillary arrays with liquid crystals in their isotropic phase, and demonstrated highly efficient optical image/beam switching and intensity/power controlling processes.				
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1.0. PROJECT SUMMARY, METHODS, AND PROCEDURES

1.1. Summary

The research program was aimed at gaining a complete fundamental understanding of recently discovered nonlinear electro-optical effects in liquid crystalline system, and exploring their applications in holographic storage, optical wave mixing and phase conjugation beam/image processing and modulation devices. In particular, the mechanisms and dynamics of photo-charge generation, space charge fields and the resultant refractive index change and holographic grating formation in dye- and fullerene doped-nematic liquid crystal have been quantitatively investigated. These work have established such liquid crystalline materials as among the most sensitive and nonlinear optical materials for performing holographic storage, wave mixing and beam and image processing and switching processes. Basic understanding of the photophysics, electrodynamics and electro-optical properties of these liquid crystalline material systems will be important for related fields such as electro-optical flat panel displays and image processing, and new material development. We have also investigated nonlinear fiber arrays made by filling capillary arrays with liquid crystals in their isotropic phase, and demonstrated highly efficient optical image/beam switching and intensity/power controlling processes.

1.2. Methods and Procedures -Technical Details

Nematic liquid crystals possess broadband (0.4 μm - 12 μm) birefringence and transparency, and unusually large susceptibility to ac, dc and optical fields. These properties, in combination with low cost, low power consumption and compatibility with semiconductor optoelectronics materials and technologies, have led to their ever increasing use in many optoelectronic display, image/data storage and processing devices. This program was focused on an extremely large nonlinear optical effect discovered in nematic liquid crystal films containing photo-charge producing dopants such as laser dyes or fullerene C60, and novel optical elements such as nonlinear fiber arrays made by filling capillary arrays with these nonlinear liquids.

1.2.1. Optically induced nematic axis reorientation and refractive index change

In analogy to reorientation by ac or static fields, an impinging optical field will realign the director axis of the liquid crystal through the dipolar interaction [1]. The process is governed by a balance equation between the applied torque and the restoring torques acting on the director axis [1]. The reorientation angle θ , and the resulting extraordinary index change $\Delta n = n_e(\beta + \theta) - n_e(\beta)$, depend on whether the interaction is transient or steady state, and on other boundary conditions[1]. In the steady state, Δn in most cases is proportional to the optical intensity I , i.e., $\Delta n = n_2 I$, where n_2 is the nonlinear coefficient. In previous studies of purely optically induced

director axis reorientation, i.e., $M_{\text{appl}} = M_{\text{op}}$, typical n_2 values obtained for 50-100 μm thick films are on the order of $10^{-4} \text{ cm}^2/\text{Watt}$ [1]. In some dye-doped nematic films [2], the optically excited dye dopant molecules exert an intermolecular torque M_m that could be much greater than the optical torque M_{op} . The nonlinear index coefficients n_2 are on the order of $10^{-3} \text{ cm}^2/\text{W}$. However, these dyed-films are highly absorptive, and invariably incur thermal effects that will present problems in practical devices.

1.2.2. Space charge fields and unusually large nonlinearities.

In our research program, we discovered that when the nematic liquid crystal is doped with photocharge producing agents such as Fullerene C_{60} , an even larger reorientational effect was observed[3-5]. The incident light generates photocharges from the dopant, which migrate and diffuse within the material and set up various dc space charge fields. Through experiments and theories [3-5], we have quantitatively identified that three major space charge fields. One is similar to those encountered in photorefractive inorganic crystals or semiconductors. For an incident optical intensity grating function of the form $I_{\text{op}} = I_0 (1 + m \cos(q\xi))$, the so-called photorefractive space charge field is given by [6] :

$$E_{\text{ph}} = qvmk_b T [(\sigma - \sigma_d) / (2e\sigma)] \cos(q\xi - \pi/2) \quad (1a)$$

where k_b = Boltzmann constant, σ = illuminated conductivity, σ_d = dark state conductivity, $v = (D_+ - D_-)/(D_+ + D_-)$, where D_+ and D_- are the diffusion constants for the positively and negatively charged ions, respectively; m is the optical intensity modulation factor, $q = 2\pi/\Lambda$ is the magnitude of the grating wave vector, and ξ is the coordinate along q .

Two other space charge fields arise from the conductive and dielectric anisotropies of the reoriented nematic liquid crystal in the presence of a small applied dc field. For a spatially varying director axis reorientation angle θ , the space charge fields are given by [3]:

$$E_{(\Delta\sigma, \epsilon)} = E_{\text{dc}} [(\sigma, \epsilon)_{//} - (\sigma, \epsilon)_{\perp} \sin 2\theta] / 2 [(\sigma, \epsilon)_{//} \sin 2\theta + (\sigma, \epsilon)_{\perp} \cos 2\theta] \quad (1b)$$

where $(\sigma_{//} - \sigma_{\perp})$ is the conductivity anisotropy and $(\epsilon_{//} - \epsilon_{\perp})$ the dielectric anisotropy, and E_{dc} is the applied dc field.

These space charge fields, in combination with the applied dc field, create a torque M_{ph} on the liquid crystal director axis and causes reorientation and large refractive index changes. In general, for a 25 μm thick film under a dc bias voltage of 1.5 V, the n_2 values are about $10^{-3} \text{ cm}^2/\text{W}$ [3]. An important feature of this effect is that it is purely orientational effect, with negligible thermal effect due to the very small amount of dopant involved. Using this new effect, we have demonstrated dynamic and storage holographies [3,5] beam amplification processes[7].

1.2.3. The most nonlinear optical material known to date

Recent studies [8] in methyl-red doped nematic films have shown that these photo-induced space charge fields can be so large that no applied dc field is needed to create observable director axis reorientation effect. This is a useful feature for practical application, as it avoids dc field-induced instabilities and dynamic scattering effects. The much larger production efficiency of photo-charges and fields in the methyl-red doped liquid crystal manifests itself in the form of a sizable dc photo-voltages across the ITO-electrode coated windows. For a particular cell thickness, the photo-voltage build-up time is dependent on the optical intensity. The build up time decreases from 1s to 100 ms as the incident laser intensity is raised from 0.3 to 3 mW/cm^2 . At 10 mW/cm^2 , the build up time drops to about 10 ms, the usual limitation of nematic liquid crystal response times. In general, the rise times also decrease for thinner samples. These dynamics are reflected in the observed response times of the nonlinear optical processes such as grating diffraction. We have observed similar effects in planar, twisted and homeotropically aligned nematic cells.

By investigating the temperature dependence of grating diffraction for two different optical intensities, we have confirmed that the nonlinear index change is due to nematic axis reorientation effect [1]. Using different pump and probe beam polarizations, we have also ruled out thermal effect for laser intensities in the 1 mW/cm^2 regime. From these grating diffraction experiments, we have estimated that the index coefficient n_2 of a methyl red doped [0.5% by weight concentration] nematic film are on the order of 6 cm^2/W [8]. With the use of a small dc or ac field, these effects can be enhanced, and n_2 can exceed 10 cm^2/W . Therefore, these methyl-red doped nematic films are arguably the most nonlinear optical material known to date. Using these films, we have demonstrated optical limiting, image conversion and visualization [9] at very low thresholds comparable to the more expensive commercial liquid crystal light valves used in similar studies [10].

1.2.4. Comparison of dye-doped nematic liquid crystals with other materials.

The potential of dye-doped nematic films for advanced applications become apparent when we make a quantitative comparison among some exemplary nonlinear optical materials[11-18]

reported in the literature. In Table 1, we list the nonlinear index coefficients n_2 of some exemplary nonlinear optical materials. Among all the nonlinear optical materials, methyl-red doped nematic films possess arguably the largest nonlinear index coefficient.

Table 1. Refractive Index Coefficients of Nonlinear Optical Materials

Material and Nonlinearities order of magnitude	n_2 (cm ² /Watt)
Nematic Liquid Crystal	
Orientational Nonlinearities	
Purely optically induced[ref.1]	10 ⁻⁴
Excited dopant assisted[ref.2]	10 ⁻³
Photorefractive -C60 doped [ref.3]	10 ⁻³
Photorefractive -methyl-red doped[ref.8]	10
GaAs bulk [ref.13]	10 ⁻⁵
GaAs MQW[ref.14]	10 ⁻³
Photorefractive crystals [ref.18]	10 ⁻⁴
CS ₂ [ref.1]	10 ⁻¹³

For image processing, where the switching speed as well as transparency of the material are important factors, an often used figure of merit is the parameter $\chi^{(3)}/\alpha\tau$. For methyl red doped liquid crystal[3], we have $\alpha = 150 \text{ cm}^{-1}$, $\tau = 10 \text{ ms}$ $\chi^{(3)} = 3.13 \times 10^{-6} (\text{m}^2/\text{V}^2)$, so $\chi^{(3)}/\alpha\tau = 209 (10^{-10} \text{ m}^3\text{V}^{-2}\text{s}^{-1})$. The values for some well known or recently developed materials are tabulated in Table 2.

Table 2. Switching Efficiency $\chi^{(3)}/\alpha\tau$ of Various Materials

Materials [ref.]	$\chi^{(3)}/\alpha\tau (10^{-10} \text{ m}^3\text{V}^{-2}\text{s}^{-1})$
PTS crystal [12]	80
GaAs Bulk [13]	30
GaAs MQW[14]	300
Cis-trans isomery[15]	0.01
Bacteriorhodopsin[16]	0.05
α 6-thiophene[17]	1
C60 [11]	0.03
methyl-red doped LC film[5]	209
C ₆₀ -doped LC film [3,5]	2

2.0. STATEMENT OF TASKS PERFORMED

The tasks completed in this three year program are summarized as follows.

2.1. Studies of photo-charge production in dye- and Fullerene C60-doped nematic film; space charge distribution and space charge field formation; dopants photochemistry.

Using standard techniques, the photo-physical properties of these films such as dielectric constants, conductivities, photocharge production efficiency, absorption and scattering losses and Freedericksz transition thresholds were measured. The dependence of the wave mixing processes on film thickness, crystalline axis alignment, and the effects of flows and instabilities have been systematically studied. These studies enable us to quantitatively determine the roles played by various optical and material parameters in holographic grating and image formation.

2.2. Studies of the interplay between the optical field and the applied dc fields in dictating the magnitude and dynamics of the director axis reorientation; investigation of nematic flows.

Paralleling the material studies outlined above, we have conducted optical wave mixing experiments to further probe the mechanisms and dynamics of liquid crystal director axis reorientations in these doped films, and to test, evaluate, and design new holographic grating storage, beam/image modulation, and switching devices. We have identified the parameter set that will optimize the nonlinear coefficient and the response times, while minimizing the required beam energy/power densities and absorption loss.

2.3. Investigation and optimization of permanent holographic grating writing and electronic modulation speeds.

From both application and basic standpoints, it is important to understand the basic processes of photo-ions (or charges) production, the resulting space charge fields and the accompanying electro-optical phenomena in these doped films. Our material characterization efforts include a systematic study of solutions of laser dyes (e.g., R6G) and fullerenes (C60 and C70) in various types of nematic liquid crystals characterized by large birefringence, electro-optics coefficient and nematic range. Specifically, we have also established the type of doped nematic films that will exhibit stable alignment and physical properties with the future development of practical storage/modulation switching devices in mind.

2.4. Liquid crystal cored nonlinear optical fiber arrays.

One approach of overcoming the thickness limitation of liquid crystal films is by fabricating fiber structures such as image transmission faceplates, using the less scattering isotropic

phase. We have discovered that Fullerene C60-doped isotropic liquid crystal (ILC) possesses high efficiencies for Reverse Saturable Absorption [RSA] and Excited State Absorption (ESA) and Two-Photon Absorption [TPA] processes to enable optical limiting of very short laser pulses. These limiting actions are characterized by eye-safe level clamped transmission and are currently being applied in developing all-optical eye/sensor protection devices in a Joint Services program.

We have also developed a quantitative theoretical model that accounts for these RSA, ESA and TPA processes. The model was successfully employed to analyze the nonlinear transmission of picosecond and nanosecond laser pulses in the liquid crystal cored fibers. These theoretical considerations and experimental results enable us to characterize the limiting effectiveness of the material by an effective optical limiting coefficient β_{eff} . For a particularly nonlinear liquid synthesized at Penn State, the β_{eff} value obtained ~ 10 cm/GW with picosecond laser pulses, and ~ 300 cm/GW with nanosecond laser pulses, which ranked largest among all the materials currently under investigation.

3.0. Conclusion

This research resulted in the identification of a new class of nonlinear electro-optical holographic grating storage and switching material that will complement existing materials in many respects, stemming from the various unique and practically useful properties of nematic liquid crystals. By fabricating versatile structures (pixeled cells, planar waveguide, fiber arrays multiple films, and solid state polymer dispersed films), one can also envision the realization of many lower-power and low bias-voltage requirement devices using the newly discovered nonlinear electro-optical effects. The program of study has also resulted in quantitative understanding of the various photophysical, electrodynamics, electro-optical and nonlinear optical properties of nematic liquid crystals. These new insights will be important for a wide range of beam/image processing, control, steering and adaptive optics applications. The discovery of the most nonlinear optical material (methyl red dye doped nematic liquid crystal) is particularly noteworthy. Using these films, we have demonstrated optical limiting at nanowatt laser power, and image conversion at $\mu\text{W}/\text{cm}^2$ intensity, which are both several orders of magnitudes lower than those obtainable from other existing nonlinear optical materials. This ushers in a host of highly promising future research and development possibilities in beam/image processing, control and adaptive optics applications.

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4.0. PUBLICATIONS AND PRESENTATIONS

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