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13. ABSTRACT (Maximum 200 words) This program is focused on understanding recent discoveries of the optical nonlinearities of the mesophases of liquid crystals, and novel nonlinear optical phenomena enabled by the extremely large nonlinearities in these material systems. Both collective and individual molecular optical properties have been investigated experimentally. Quantitative theories of the basic physics as well as novel phenomena and the working principles of multifunctional optical devices have also been developed. Specifically, (i) the large optical nonlinearities of nematic liquid crystals in the optical communication wavelength regime (1.55µm) as well as the visible region have been quantitatively established. (ii) All-optical self-action processes such as stimulated scattering and polarization switching, and self-starting optical phase conjugation using thin (microns) nematic films with very low power cw infrared lasers have been demonstrated, along with the development of quantitative theories. (iii) Novel optical elements/devices such as liquid-crystal photonic crystals, nonlinear optical fibers and fiber arrays were developed using nematic as well as isotropic phase liquid crystals. (iv) Quantitative theoretical descriptions of their tunable and nonlinear transmission/reflection properties, and optical limiting actions and practical devices; feasibility demonstration of the nonlinear fiber array limiting devices. (v) Demonstration of the feasibility of optical soliton formation in nematic liquid crystals. (vi) Synthesis of a new class of extremely nonlinear liquid with superior optical limiting performance, wide temperature stability and very fast (picosecond – nanosecond) nonlinear electronic absorption properties suitable for limiting applications against agile frequency lasers. These newly developed theories and experimental feasibility demonstration will find applications in various compact, low-power-consumption, light-weight versatile optical switching, sensor protection and image/signal processing photonic devices.				
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Final Report

Author: Iam Choon Khoo

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1. Statement of Problems Studied.

The program of experimental and theoretical studies is centered on discoveries made in the principal investigator's laboratory of nematic liquid crystalline systems that exhibit record breaking high optical nonlinearities and photosensitivity. These unique combination of nonlinear optical and optoelectronics properties were observed in nematics doped with photo-charge producing compounds such as methyl-red dye, or photochromic materials such as azobenzene liquid crystals. The program objectives are to elucidate and completely characterize the physical origins of such supra nonlinearities, and to explore the feasibility of utilizing these unique materials in advanced holographic, optical modulation, mixing, limiting and adaptive optics devices. Well established experimental techniques such as holographic grating diffraction, stimulated scattering and optical phase conjugation and pump-probe spectroscopy were employed to investigate the dynamics and parametric dependence of the nonlinear optical responses in these complex material systems, using lasers of wide temporal and spectral characteristics. Paralleling these experimental studies, theoretical formalisms for the photo-induced space charge field formation, isomerization induced order parameter change, crystalline axis reorientation and refractive index modulation, and cross-polarized wave mixing processes in stimulated orientation scattering processes were also made. All the projects proposed in the research program were successfully completed. Some device feasibility demonstrations enabled by such extraordinarily large nonlinearities were also conducted. These newly developed theories and experimental feasibility demonstration will find applications in various compact, low-power-consumption, light-weight versatile optical switching, sensor protection and image/signal processing photonic devices.

2. Scientific Progress and Accomplishment

In this research program, we have made the following noteworthy accomplishments:-

(i) Discovery of a class of special dopant modified nematic liquid crystals (NLC) that possess by far the largest *effective* nonlinear index coefficient

(ii) The large optical nonlinearities of nematic liquid crystals in the optical communication wavelength regime ($1.55\mu\text{m}$) have also been quantitatively established.

(iii) All-optical self-action processes such as stimulated scattering and polarization switching, and self-starting optical phase conjugation using thin (microns) nematic films with very low power cw infrared lasers have been demonstrated, along with the development of quantitative theories.

(iv) Novel optical elements/devices such as liquid-crystal photonic crystals, nonlinear optical fibers and fiber arrays were developed using nematic as well as isotropic phase liquid crystals.

(v) Quantitative theoretical descriptions of tunable and nonlinear transmission/reflection properties of photonic and optical limiting actions of the photonic crystal waveguides and nonlinear fiber array devices.

(vi) Demonstration of the feasibility of optical soliton formation in nematic liquid crystals.

(vii) Synthesis of a new class of extremely nonlinear liquid with superior optical limiting performance, wide temperature stability and very fast (picosecond – nanosecond) nonlinear electronic absorption properties suitable for limiting applications against agile frequency lasers.

These accomplishments are all documented in published refereed journal articles, proceedings of technical conferences, and master and Ph. D. student thesis as well as invention disclosures. In the next few sections, some of these scientific findings are highlighted.

2.1 Extremely Nonlinear liquid crystalline materials.

Liquid crystals exist in many mesophases intermediate between the isotropic(liquid) state and the crystalline phases. From the point of view of optoelectronics and nonlinear optics, the nematic phase stands out as the most important one. They possess unusually large birefringence (n_e-n_o can be as large as 0.7), and broadband (near UV to far infrared) transparency. Unlike their liquid- or crystalline- phase counterparts, where it takes a very strong field to reorient the molecules, nematic liquid crystals are characterized by an easy susceptibility of the molecular axis to perturbation by an external field. In the optical domain, this translates to an extremely nonlinear optical response.

Liquid crystals are also extremely nonlinear at the individual molecular level, as a result of the constituent organic molecules' electronic structures. In their liquid phase, these materials can be easily incorporated into novel optical structures/elements for applications where nanosecond and faster speeds are required.

In this research program, we have discovered and established the basic science of a class of special dopant modified nematic liquid crystals (NLC) that possess an *effective* nonlinear index coefficient $n_2 \gg 1 \text{ cm}^2/\text{W}$ (n_2 is defined by index change $\Delta n = n_2 I$, where I is the optical intensity in W/cm^2). In Tables 1 –2 we list the *effective nonlinear index coefficients* n_2 and *switching efficiency* $\chi^{(3)}/\alpha\tau$ of these NLC materials along with other well known classes of nonlinear optical materials^[25-31]. Note that $\chi^{(3)}$ is the equivalent third order nonlinear susceptibility; α = absorption constant and τ is the response time. We also insert the corresponding data for commercial [and much more costly] Optically Addressed Liquid Crystal Spatial Light Modulator [LC-OASLM] by estimating its index changing efficiency under a given illumination optical intensity. From this standpoint of possessing $n_2 \gg 1 \text{ cm}^2/\text{W}$, LC-OASLM is a 'supra-nonlinear' optical device, enabled by incorporating a highly photosensitive photo-conducting semiconductor layer. The supra-nonlinear LC materials discovered in this program clearly stand out as rather promising candidates for further investigations and application possibilities.

Table 1. Refractive Index Coefficients of Some Nonlinear Optical Materials

Materials	Order of Magnitude of n_2 (cm^2/W)
Nematic Liquid Crystal	
Purely optically induced	10^{-4}
Photorefractive -C60 doped	10^{-3}
Photorefractive-methyl-red doped	10
BMAB doped NLC	>2
C60/nanotube doped film	>20
OASLM – LC	<i>estimated</i> ~ 10
GaAs bulk	10^{-5}
GaAs MQW	10^{-3}
Photorefractive crystals/polymers	10^{-4}
Bacteriorhodopsin	10^{-3}
Cis-trans isomery	10^{-3}

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

Materials [ref.]	$\chi^{(3)}/\alpha\tau$ ($10^{-10} \text{ m}^3\text{V}^{-2}\text{s}^{-1}$)
GaAs Bulk	30
GaAs MQW	300
Bacteriorhodopsin	0.05
Cis-trans isomery	0.01
methyl-red doped LC film	200
C60/nanotube doped film	~500
OASLM - LC	estimated ~200

Note: $n_2 = 0.105 \times \chi^{(3)}_{\text{cgs}} / n_0^2$ [cm^2/Watt]; $\chi^{(3)}$ [in m^2/V^2] = $1.39 \times 10^{-8} \chi^{(3)}_{\text{cgs}}$ [in esu]. For MRNLC, $\alpha = 150 \text{ cm}^{-1}$, $\tau = 10 \text{ ms}$, $\chi^{(3)} = 3.13 \times 10^{-6} (\text{m}^2/\text{V}^2)$, so $\chi^{(3)}/\alpha\tau = 209 (10^{-10} \text{ m}^3\text{V}^{-2}\text{s}^{-1})$

2.2. Novel Nonlinear Optical Phenomena

We have also made several first time studies of novel nonlinear optical phenomena with practical application potentials. These processes are characterized by very low operation optical power ($\mu\text{Watt} - \text{mWatt}$) and applicability over very broad spectral and temporal ranges. For example, using methyl-red doped nematic liquid crystalline [MRNLC] films, we have demonstrated a very large dynamic range all-optical anti-laser jamming device capable of removing glares and laser dazzle, c.f., Fig. 1a. The same MRNLC films were also used in feasibility demonstrations of all-optical image processing operations such as wavelength conversion, contrast reversal, c.f. Fig. 1c, image addition and subtraction, and fabrication of tunable liquid crystal 2-D photonic crystal structures. In collaboration with others, we have also demonstrated transient and storage holographic applications with these films; it is possible to optically write holographic gratings with resolution as high as 1000 lines/mm, that can be switched at fairly high speed [$<\text{ms}$] switching using dual frequency drive.

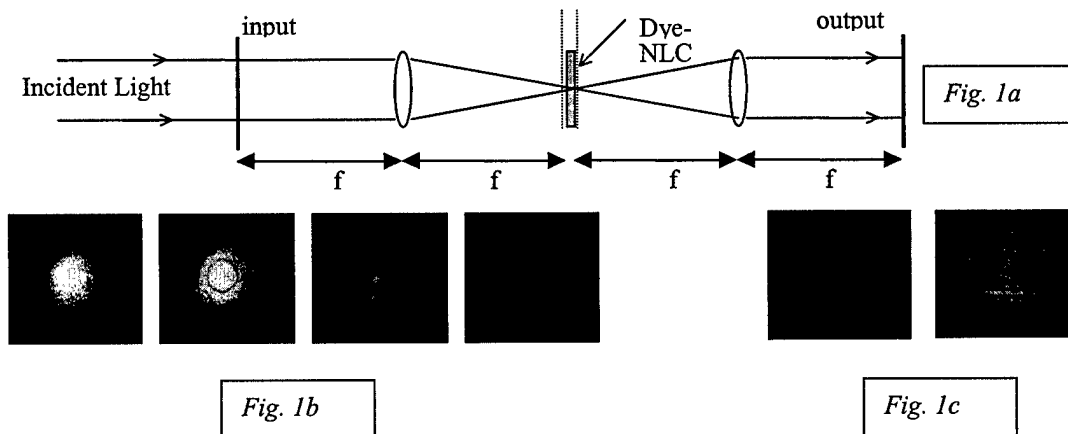
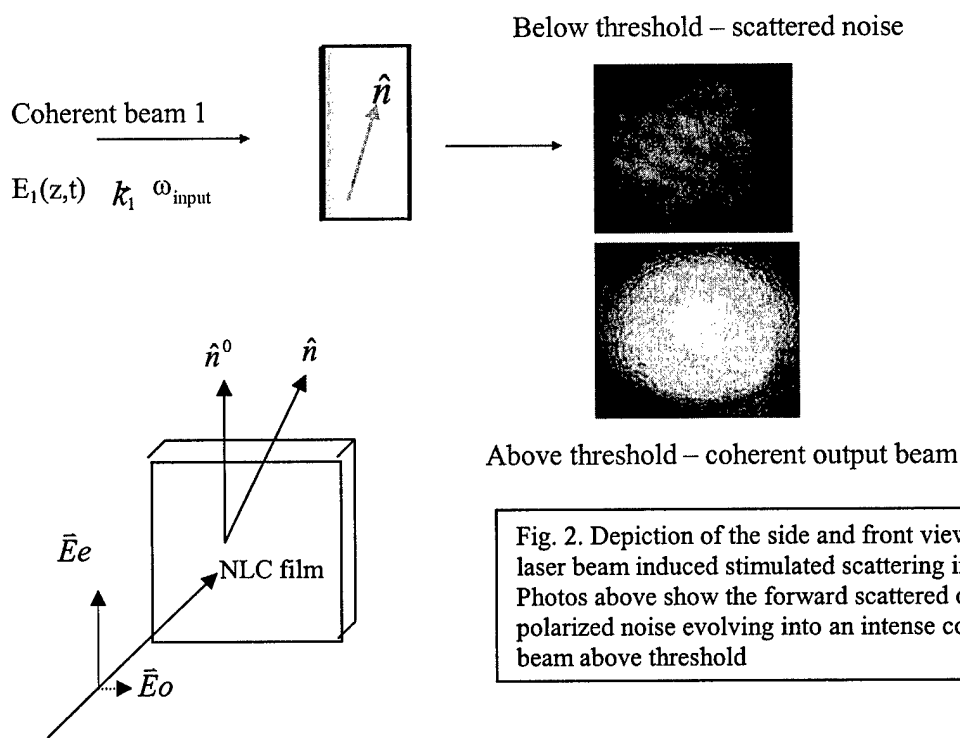


Fig. 1a. Schematic of NLC film in a 4-F optical system for all-optical image processing. Fig. 1b Photos showing anti-laser jamming action - the transmission through the LC film is reduced to almost vanishing value the as incident laser intensity is increased - in the order from left to right. Switched off can be in 10^{-6} s using supra-nonlinear azo-doped liquid crystal or MRNLC. Fig. 1c. Image contrast reversal by optically induced self-phase modulation with the NLC film.

Perhaps the best illustration of the usefulness of LC's large broadband nonlinearity is stimulated orientation scattering (SOS), in which a scattered noise [lower] frequency component from an incident coherent beam experiences unidirectional energy flow from the input beam via two-beam coupling effect, in a manner analogous to stimulated Brillouin scattering (SBS), c.f. Fig. 2. In SBS, the phase matching is provided by the sound wave [a travelling density grating], and the process usually requires very high laser power [MW in cm-long liquid column] and/or very long interaction length [cm's to meters]. In liquid crystal, the unidirectional energy transfer involved in SOS is mediated by a dynamic grating formed by mixing of an input 'pump' beam with its scattered noise, c.f. Fig. 2. The process requires very low optical power (mW) in very short (100's micron) interaction length.



Another important feature of stimulated scattering is the non-resonant nature of the underlying optically induced director axis reorientation process. For two-wave mixing between the incident e-wave and its scattered o-wave noise, the noise component that is Stoke-shifted by $\Delta\omega \sim \Gamma^{-1}$ (the inverse of the relaxation time constant) will experience maximal gain and grow to a coherent beam.

The maximum gain coefficient $G_m (cm \cdot W^{-1}) = \frac{(n_e + n_o)^2 \lambda}{8\pi c n_e K_2}$ shows a 'weak' dependence on the

wavelength. Since n_e and n_o do not change appreciably throughout the visible-near IR regime, the nonlinear wave mixing process will occur with similar efficiency throughout, as demonstrated in our recent studies with 1.55- μm laser and visible laser, c.f. Fig. 3.

For application in the infrared, it is important to point out here that for NLC's, their scattering loss scales inversely as the laser wavelength (loss $\sim \lambda^{-n}$ ($n \geq 2$)). This means one could use thicker sample and obtain higher efficiency. Nematic liquid crystals are therefore important addition to the relatively limited availability of nonlinear optical materials in this important infrared wavelength regime.

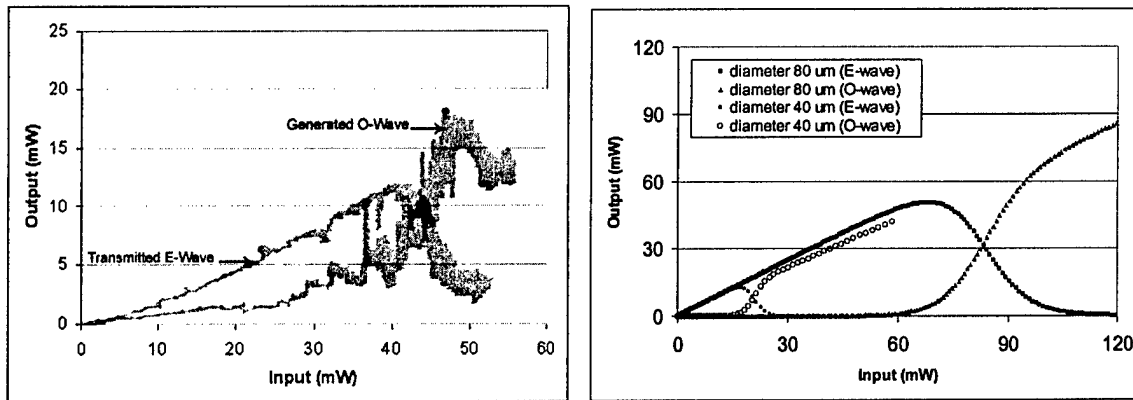


Fig. 3 (a) Experimentally obtained output e- and o-wave as functions of input laser power for a cw $1.55 \mu\text{m}$ laser. (b) Calculated output e- and o-wave as functions of input e-wave power.

2.3 Isotropic Phase LC fiber array and limiting action

The isotropic(liquid) phase of many liquid crystals also presents important pleasant surprises. We have discovered that the constituent molecules of some liquid crystals possess fast acting and broadband [*sub-picosecond response times*] Two-Photon Absorption (TPA) and Excited State absorption(ESA) properties. Because of their fluid nature, these liquid-phase liquid crystals can be easily incorporated into capillary arrays to form nonlinear fiber arrays, c.f. Fig. 4a. The fiber array functions as high quality transmission faceplate for low light level images; on the other hand, the nonlinear absorption properties of the core liquid would 'clamp' the transmission of high intensity laser pulses, c.f. Fig. 4b; agile frequency visible laser pulses ranging in power/intensity over 3 orders of magnitude [from μJ to $\gg \text{mJ}$ in energy] can be passively 'limited' to below the eye/sensor safe level [$< 1 \mu\text{J}$], c.f. Fig. 5.

This self-action effect can also be applied to pulse shaping and stabilization control. We are currently developing nonlinear fiber core liquids capable of withstanding very large temperature variation [from -40°C to $>100^\circ\text{C}$].

These nonlinear fiber arrays possess several *advantageous properties* compared to thin films or multiple tandem film devices as the latter suffer from limited [small] field-of-view. For nonlinear fiber arrays, the *field of view* can be very large [over 45°]. Another advantage of such fiber device is the extension of the nonlinear interaction region by the waveguiding property of the fiber core. In conjunction with an *opaque cladding*, this extended waveguiding nonlinear core region enable further blocking of the nonlinear scattered light from going to downstream optics, resulting in *low clamped transmission* [$< 1 \mu\text{J}$ encircled energy] for input $> 1 \text{ mJ}$ and a *large dynamic range* [$>10^3$]. Furthermore, the *nonlinear liquid* layer at the entrance plane of the fiber array possesses *self-healing* property. Upon irradiation by intense laser pulses, the 'damaged' liquid region vaporizes and the bubble thus formed floats away. Moreover, the bubble also scattered the incident laser strongly, and contribute to further lowering of the transmission.

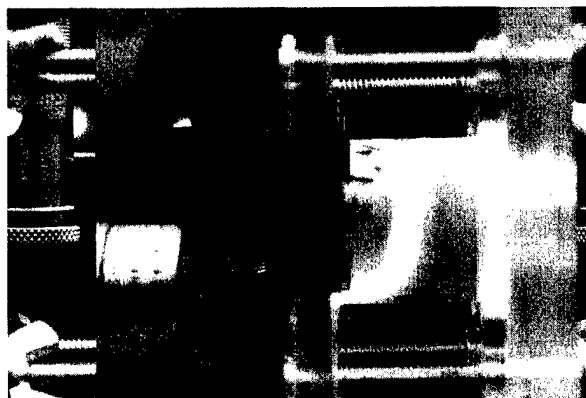
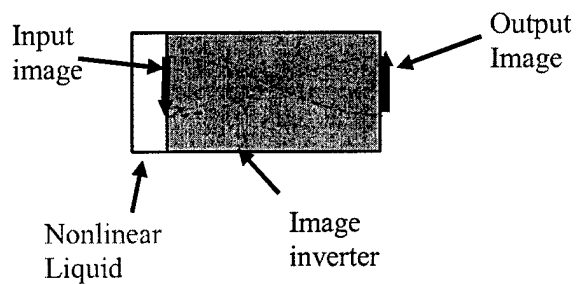
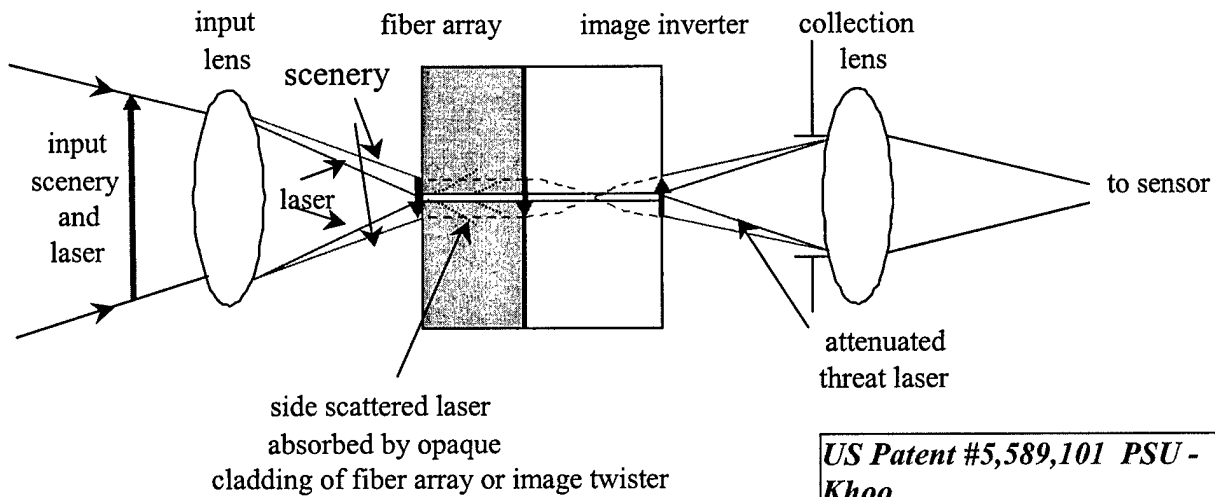
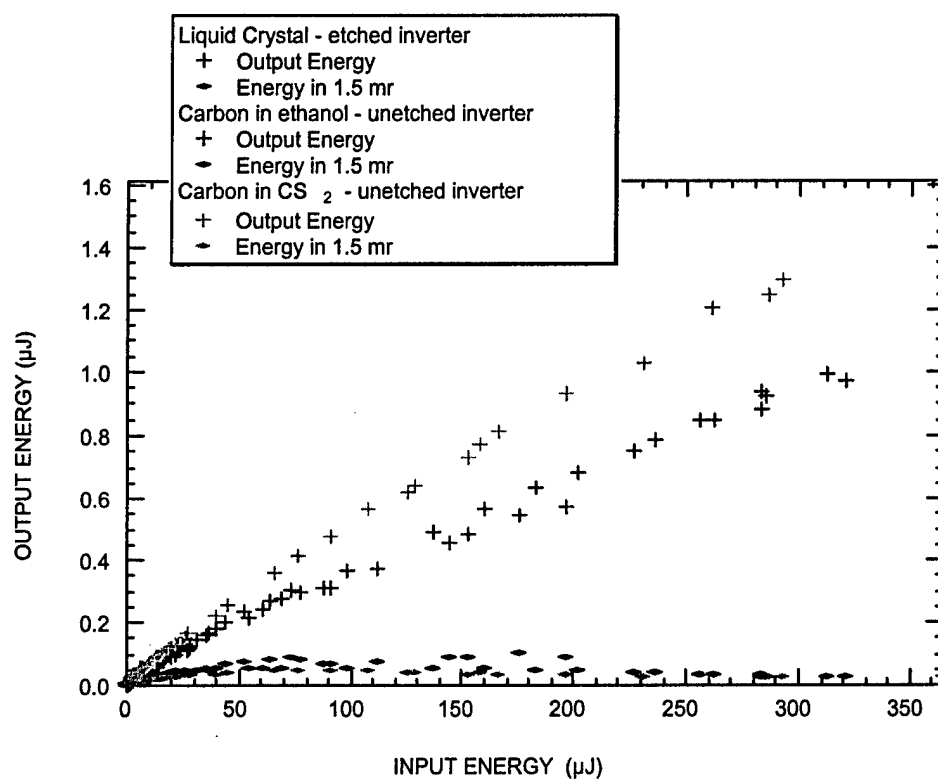


Fig. 4. Schematic depiction of the nonlinear fiber array for imaging and passive optical limiting application. Insert shows an alternative configuration using the liquid developed at PSU in conjunction with a fiber image inverter. Photograph below shows side view of the limiter [liquid+fiber image inverter] with an input laser from the left.



Limiting with liquid crystal material compared to carbon suspensions

Fig. 5. Some optical limiting results obtained with isotropic-phase liquid crystal filled fiber array compared with other materials/device configurations.

3. List of Journal Papers Published

1. M. Y. Shih, I. C. Khoo, A. Shishido, M. V. Wood, P. H. Chen, "All-Optical Image Processing with A Supra Nonlinear Dye-Doped Liquid Crystal Film", *Opt. Letts.* 25, 978-980 (2000).
2. M. Peccianti, A. De Rossi and G. Assanto, A. De Luca and C. P. Umeton and I. C. Khoo, "Electrically assisted self-confinement and waveguiding in planar nematic liquid crystal cells," *Appl. Phys. Letts.* 77, pp 7-9 (2000).
3. I. C. Khoo, M. Y. Shih, M. V. Wood and P. H. Chen' "Extremely nonlinear photosensitive nematic liquid crystal film," *Synthetic Metals*, 7413, 1-6 (2000)
4. I. C. Khoo and Y. Liang, "Stimulated Orientational and Thermal Scatterings and Self-Starting Optical Phase Conjugation with Nematic Liquid Crystals," *Phys. Rev. E* 62, pp 6722 -6733 (2000)
5. I. C. Khoo, P. H. Chen, M. Y. Shih, A. Shishido, S. Slussarenko, "Supra Optical Nonlinearities of Methyl-Red and Azobenzene Liquid Crystal -doped Nematic Liquid Crystals," *Mol. Cryst. Liq. Cryst.* 358, 1-13 [2001]

5. I. C. Khoo, M. Y. Shih, A. Shishido, P.H. Chen and M. V. Wood , "Liquid Crystal Photorefractivity – Towards Supra-Optical Nonlinearity," Invited Paper *Opt. Mate.* 18, 85-90 (2001).
7. I. C. Khoo, M. Y. Shih and A. Shishido, "Supra Optical Nonlinearities of Photosensitive Nematic Liquid Crystals," *Mole. Cryst. Liquid. Cryst.* 364, 141-149, (2001).
8. Atsushi Shishido, Ivan B. Diviliansky, I. C. Khoo and Theresa S. Mayer Suzushi Nishimura, Gina L. Egan, Thomas E. Mallouk, "Direct Fabrication of Two-Dimensional Titania Arrays using Interference Photolithography," *Appl. Phys. Letts.* 79, 3332-3334 (2001).
9. M. Y. Shih, A. Shishido and I. C. Khoo, " All-optical image processing by means of photosensitive nonlinear liquid crystal film: edge enhancement and image addition/subtraction," *Opt. Letts.* 26, pp 1140-1142 (2001).
10. I. C. Khoo, A. Diaz, M. V. Wood and P. H. Chen , "Passive optical limiting of picosecond – nanosecond lasers using highly nonlinear organic liquid cored fiber array," *IEEE J. Selected Topics in Quantum Electronics*, Vol. 7, no. 5, pp. 760-768 (2001).
11. Ivan B. Diviliansky, Atsushi Shishido, I. C. Khoo, Theresa S. Mayer, David Pena, Suzushi Nishimura, and Thomas E. Mallouk, "Fabrication of two-dimensional photonic crystals using interference lithography and electrodeposition of CdSe," *Appl. Phys. Letts.* 79, 3392-3394 (2001).
12. I. C. Khoo, M. Kaczmarek, M. Y. Shih, M. V. Wood, A. Diaz, J. Ding and Y. Zhang, "Nonlinear optical effects in nematic liquid crystal films in the 1.55 μ m spectral region," *Mol. Cryst. Liq. Cryst.* 374, pp. 315-324 (2002).
13. M. Peccianti, G. Assanto, A. De Luca, C. Umeton, M. A. Karpierz, and I. C. Khoo: Light Self-Confinement in Planar Cells with Nematic Liquid Crystals *J. Communications Technology and Electronics* , 47(7), 790 (2002).
14. I. C. Khoo, J. ding, A. Diaz, Y. Zhang and K. Chen, "Recent studies of optical limiting, image processing and near-infrared nonlinear optics with nematic liquid crystals," *Mol. Cryst. Liq. Cryst.* 375, pp. 33-44 (2002).
15. G. Assanto, M. Peccianti, C. Umeton, A. De Luca and I. C. Khoo, "Coherent and incoherent spatial solitons in bulk nematic liquid crystals," *Mol. Cryst. Liq. Cryst.* 375, pp 617-629 (2002).
16. Malgosia Kaczmarek, Min-Yi Shih, Roger S. Cidney and I. C. Khoo, "Electrically tunable, optically induced dynamic and permanent gratings in dye-doped liquid crystals", *IEEE J. Quantum Electronics* 38, pp 451-457 (2002).
17. I. C. Khoo and J. Ding, "All-optical cw laser polarization conversion at 1.55 micron by two beam coupling in nematic liquid crystal film," *Appl. Phys. Letts.* 81, pp. 2496-2498 (2002).

4. Invited Conference Presentations

- *1. I. C. Khoo. "Liquid Crystal Photorefractivity – Towards Supra-Optical Nonlinearity," Invited Paper. European Material Research Symposium, Strasbourg, France, 5/2000.
- *2. I. C. Khoo, "Nonlinear Photonics of Isotropic Liquid and Nematic Liquid Crystal for Optical Limiting Application," Invited Paper. Second Int. Conference on Optical Power Limiting, Venice, Italy 7/2000.
- *3. I. C. Khoo, "Supra Optical Nonlinearities of Photosensitive Nematic Liquid Crystals," Invited Paper. 2000 Int. Liquid Crystal Conference, Sendai, Japan, 7/2000.

- *4. I. C. Khoo, "Modeling Nonlinear Photonics of Nonlinear Fiber Core for Optical Limiting Application" Invited Paper. SPIE SPIE's Int. Symp. on Optical Science, Engineering and Instrumentation, San Diego, 8/2000.
- *5. I. C. Khoo, "Photorefractive and Supra Nonlinear Nematic Liquid Crystals for Optical Switching and Image Processing Application," 1st International Conference on Molecular Synchronization for Design of New Materials," Tokyo, Japan, 9/2000.
- *5. I. C. Khoo, "Supra-nonlinear nematic liquid crystal for dynamic and storage holography and image processing", Optical Society of America Annual Meeting, Providence, Rhode Island, 10/2000
- *7. I. C. Khoo, "Novel nonlinear optics and applications with liquid crystals", International Chinese Liquid Crystal Conference, Tainan, Taiwan, 12/2000.
- *8. I. C. Khoo, "Nonlinear nematic liquid crystals and phototunable 2-and 3-D photonic crystal for visible-1.55 micron spectral region," Invited Paper, Int. Conference on Frontiers of Polymers and Advanced Materials, Recife, Brazil, March 4-9, 2001.
- *9. I. C. Khoo, "Novel nonlinear optics of liquid crystals," Invited Paper – 5th International Topical Meeting on Novel Optical Materials and Applications, Cetraro, Italy, May 21-26, 2001
- *10. I. C. Khoo et al , "Nonlinear liquid crystals in periodic structures," Invited paper. SPIE International Symposium on Optical Science and Technology – Liquid Crystal V, 7/2001 San Diego.
- *11. I. C. Khoo, "Recent studies of optical limiting, image processing and novel nonlinear optics with nematic liquid crystals," Invited paper, 9th International topical Meeting on Optics of Liquid Crystals, Sorrento, Italy, 10/2001.
- *12. I. C. Khoo, "Photosensitive nematic liquid crystals for intelligent photonics," Invited paper. International Symposium on Organic electronic and Photonic Materials and Devices, Osaka, Japan 11/2001.
- *13. I. C. Khoo, Yana Zhang, A. Diaz, J. Ding, I. B. Divliansky, Kito Holliday, T. S. Mayer, V. Crespi, D. Scrymgeour, V. Gopalan, "Widely tunable nonlinear liquid crystal-based photonic crystals," Invited Paper - Liquid Crystals VI –SPIE International Symposium on Optical science and Technology, Seattle, Washington July (2002).
- *14. I. C. Khoo, "Nonlinear photonics of liquid crystals," Invited paper. International Conference on Modern Optics and Applications, Bandung, Indonesia July 3-5 (2002).
- *15. I. C. Khoo, "Photorefractivity in liquid Crystalline Materials," Invited paper - 'Sturgeon Memorial Lecture' – Cambridge University.- 2003 British Liquid Crystal Society Meeting, April 9, 2003.

5. Invention Disclosure.

5.1. I. C. Khoo, "*All-optical polarization rotation, switching, and optical limiting device for broadband [0.4 micron -1.5 micron] application*". PSU Invention Disclosure #2002-2692. Filed on August 8, 2002.

5.2. I. C. Khoo, "Fiber Array with Two-Photon+Reverse-Saturable Absorbing core for optical limiting and eye/sensor protection against agile frequency short-pulsed lasers". PSU Invention Disclosure #2002-2707. Filed on Oct. 8, 2002

6. Technology Transfer

6.1. *Navy Air Warfare Center*, Patuxent River, Maryland – Development of Helmet-mount eye/sensor protection against short-pulsed agile frequency laser.

6.2. *Wright Patterson Air force Base SBIR Phase II & Army Research Office SBIR Phase I* - Next generation liquid crystal spatial light modulator and holographic image processing films, and also low threshold cw-light source optical limiting devices.

7. Participating Scientific Personnel

The research program has involved the principal investigator and 6 graduate students [Michael Wood, P. H. Chen, M. Y. Shih, K. Chen and J. Ding].

8. Graduate theses:

Three of the students [M. Wood, M. Y. Shih and P. H. Chen] have graduated with Ph. D. degrees.