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Report Title

Extreme Nonlinear Optics with Liquid Crystals

ABSTRACT

A multi-prong experimental and theoretical studies of recently discovered extremely nonlinear liquid crystals and novel nonlinear optical processes have been conducted. Specifically, we have successfully carried out the following projects: (i) Synthesis of nonlinear nematic liquid crystals containing highly photosensitive/photoconductive nano-particulates (Gold nano-wires, CdSe nanorods); (ii) Synthesis of nonlinear liquids with large two- and multi-photon absorption and excited-state absorption coefficients; (iii) Complete characterization of the fundamental mechanisms responsible for the supra- nonlinearities of these liquid crystalline systems (iv) Design/fabrication of nonlinear fiber array and tunable 2-and 3-D liquid crystalline photonic crystals, frequency selective surfaces and core-shell nano-spheres dispersed bulk liquid crystalline film (v) Complete quantitative studies of self-action effects such as all-optical switching, optical limiting, nonlinear guided waves using visible as well as infrared lasers. Paralleling these studies, we have successfully demonstrated the feasibility of several unique and high performance holographic, modulation, mixing, limiting, adaptive optics and various self-action or optically activated devices. These devices are operational at low power/intensity levels over a wide spectral and temporal bandwidth, and have opened up new avenues for future research and development possibilities, especially when coupled to nano-structures and nano-metamaterials of unique optical, electro-optical and nonlinear-optical properties.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

1. I. C. Khoo, J. Ding, Y. Zhang, K. Chen and A. Diaz, "Supra-Nonlinear Photorefractive Response of Single-wall Carbon Nanotube- and C60-Doped Nematic Liquid Crystals," *Appl. Phys. Letts.* 82, pp. 3587-3589 (2003).
2. I.C. Khoo and A. Diaz, "Nonlinear dynamics in laser polarization conversion by stimulated scattering in nematic liquid crystal films," *Phys. Rev. E* 68 pp 042701-1 to -4 [2003]
3. I. C. Khoo, K. Chen and A. Diaz, "All-Optical Neural-net like Image Processing with Photosensitive Nonlinear Nematic Film," *Optics Letts.* 28, pp 2372-2374 [2003].
4. Y. Ye, T. S. Mayer, I. Khoo, I. B. Divliansky, N. M. Abrams and T. E. Mallouk, "Self- assembly of three-dimensional photonic crystals with air-core line defects," *J. Mater. Chem.* 12, 3637 (2002).
5. O. Ruzak, N. Collings, W. A. Crossland, T. D. Wilkinson, A. B. Davey and I. C. Khoo, "Dynamic Holographic Gratings in Methyl Red-Doped Nematic Liquid Crystals," *J. Non. Opt. Phys. & Mat.* Vol 12, pp441-446 (2003).
6. I.C. Khoo, Andres Diaz and J. Ding, "Nonlinear-absorbing Fiber Array for Large Dynamic Range Optical Limiting Application against Intense Short Laser Pulses," *J. Opt. Soc. Am B* 21, pp. 1234-1240 [2004].
7. Y. Ye, J. Ding, D. Jeong, I. Khoo and Q. Zhang, "Finite-size effect on one-dimensional coupled-resonator optical waveguides" *Phys. Rev. E* 69, 056604 (2004).
8. Malgosia Kaczmarek, Andrey Dyadyusha, Sergei Slussarenko and I.C. Khoo, "The role of surface charge field in two-beam coupling in liquid crystal cells with photoconducting polymer layers," *J. Appl. Phys.* 96, pp. 2616-2623 [2004].
9. Iam Choon Khoo, Jianwu Ding, and Andres Diaz, "Dynamics of Cross-polarization Stimulated Orientation Scattering in Nematic Liquid Crystal Film," *J. Opt. Soc. Am. B* 22, pp. 844-851 [2005]
10. Jae-Hong Park, Iam Choon Khoo, Chang-Jae Yu, Min-Sik Jung and Sin-Doo Lee, "Formation of binary phase gratings in photopolymer-liquid crystal composites by a surface-controlled anisotropic phase separation," *Appl. Phys. Letts.* 86, pp 021906-1 - 3 [2005].
11. I. C. Khoo, Yana Zhang Williams, B. Lewis and T. Mallouk, "Photorefractive CdSe and gold nanowire-doped liquid crystals and polymer-dispersed-liquid-crystal photonic crystals," *Mol. Cryst. Liq. Cryst.* 446: 233-244 (2005).
12. Jae-Hong Park and Iam Choon Khoo, "Liquid crystal beam steering device with a photopolymer prism," *Appl. Phys. Letts* 87, 091110- 091112 [2005].
13. I. C. Khoo, Kan Chen and Y. Zhang Williams, "Orientational Photorefractive Effect in undoped and CdSe Nano-Rods doped Nematic Liquid Crystal – Bulk and Interface Contributions," *IEEE J. Selected Topics in Quantum Electronics* JSTQE 12 (3), pp. 443-450 [2006].
14. E. Graugnard, J. S. King, S. Jain, C. J. Summers, Y. Zhang-Williams and I. C. Khoo, "Electric field tuning of the Bragg peak in large-pore TiO₂ inverse shell opals," *Phys. Rev. B* 72, 233105 (2005).
15. Jae-Hong Park and Iam Choon Khoo, "Liquid crystal beam deflector with a photopolymer prism," *Molecular Crystal Liquid Crystal* 454, pp.135-143 (2006).
16. I. C. Khoo, Yana Williams, Andres Diaz, Kan Chen, J. Bossard, D. Werner, E. Graugnard and C. J. Summers, "Liquid-Crystals for optical filters, switches and tunable negative index material development," *Molecular Crystal Liquid Crystal* 453, pp.309-319 (2006).
17. I. C. Khoo, D. H. Werner, X. Liang, A. Diaz and B. Weiner, "Nano-sphere dispersed liquid crystals for tunable negative-zero-positive index of refraction in the optical and Terahertz regimes," *Optics Letts.* 31, 2592 (2006)

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(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

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(c) Presentations

***Invited**

1. I. C. Khoo, "Nonlinear Optics of the Mesophases of Liquid Crystals for Optical Power Limiting applications," 3rd International Symposium on Optical Power Limiting, Sedona, Arizona, September 28-Oct. 2 (2003).
4. I. C. Khoo, K. Chen, A. Diaz, J. Ding and Yana Zhang, "All-Optical Neural-net like Image Processing with Local and Nonlocal Nonlinear Optical Thin Films" Lasers and Electro-Optics Society Annual Meeting, Tucson, AZ Oct. 27-31 (2003)
- *5. I. C. Khoo, "Nonlinear Optics of the Mesophases of Liquid Crystals" International Conference on Organic Nonlinear Optics, Serak, Korea, November 3-9 (2003).
6. S. Yin, Z. Liu, I. C. Khoo, K. W. Chung, Y Yang, "Tunable photonic crystal fibers and their applications," American Ceramic Society Meeting on Optical Waveguides: Unconventional Approaches and Applications, October 2003, Corning, NY.
- *7. I. C. Khoo, "Ultra-large Dynamic Range Nonlinear Absorption Materials" Int. Conference on Photoresponsive Organics and Polymers, Busan, Korea, Feb. 16- 20 (2004).
8. Jianwu Ding, Iam Choon Khoo, "Orientational photorefractive effect in photo-sensitive-dye doped nematic liquid crystals", Frontiers in Optics 2004 (The 88th OSA annual meeting), Rochester, NY, October 2004.
- *9. I. C. Khoo, "Nonlinear molecular photonics and controlled laser transmission through a nonlinear fiber array," SPIE European Symposium on Optics and Photonics for Defense and Security, London, Oct. 2004
10. Kan Chen, Iam-Choon Khoo, "Interface Induced Effect in Liquid Crystal Photorefractive Nonlinearity", Frontiers in Optics 2004, Rochester, NY, October 2004.
- *11. I. C. Khoo, "Liquid crystals-Photonic crystals" Invited paper. Annual Meeting of the IEEE-Lasers and Electro Optics Society, Nov. 8, 2004, Puerto Rico
12. I. C. Khoo, "All-Optical Self-Polarization Switching via two-wave mixing in nematic Liquid Crystals – from visible to infrared", Annual Meeting of the IEEE-Lasers and Electro Optics Society, Nov. 11, 2004, Puerto Rico.
- *13. I. C. Khoo, "Holographic fabrication of polymer dispersed liquid crystal photonic crystals," Invited paper, Materials Research Society Fall Meeting, Boston, Dec. 2, 2004
- *14. I. C. Khoo, "Liquid Crystals Ultra Broadband All-Optical Devices", 8th Int. Conference on Frontiers of Polymers and Advanced Materials, Cancun, Mexico April 23-28, 2005.
- *15. I. C. Khoo, "Fundamentals and Applications of Ultra-Broadband Liquid-Crystal All-Optical and Electro-Optical Properties and Processes," Int. Conference on Photonics, Biophotonics and Nanophotonics, Nanjing, China May 14-19, 2005.
- *16. I. C. Khoo, "Electro- and Nonlinear –optical Properties of Liquid Crystals," 7th Int. Topical Meeting on Novel Optical Materials and Applications," Cetraro, Italy. May 29 – June 4, 2005.
17. Andres Diaz, J. Ding, Kan Chen and I. C. Khoo, "Nonlinear dynamics of stimulated cross-polarization scattering in a nematic liquid-crystal film," in 7th Int. Topical Meeting on Novel Optical Materials and Applications," Cetraro, Italy. 6/2005.
18. Yana Williams, Kan Chen, Jay Hong Park, I. C. Khoo, B. Lewis and T. Mallouk, "Photorefractive effect in semiconductor nanorod doped liquid crystal," 7th Int. Topical Meeting on Novel Optical Materials and Applications," Cetraro, Italy. 6/2005.
- *19. E. Graugnard, J. S. King, D. Gaillot, C. J. Summers, Y. Zhang-williams and I. C. Khoo, "Liquid crystal infiltration of template patterned 3-D photonic crystals," Invited Paper in Conference on Tuning the Optical Response of Photonic Bandgap Structures II – SPIE Symposium on Optics and Photonics, San Diego, California, 8/2005.
20. I. C. Khoo, Jay Hong Park, Yana Z. Williams, Kan Chen and Andres Diaz, "Liquid Crystals for switching, frequency filters and tunable negative index material development", in Optics of Liquid Crystals conference," Clearwater, Florida, 10/2005.
- *21. I. C. Khoo and A. Diaz, "Self-action cw-pulsed lasers and glares attenuators," Invited paper, IEEE-Lasers and Electro-Optics Society Annual Meeting, Sydney, Australia [10/2005].
22. Elton Graugnard, Davy P. Gaillot, Jeffrey S. King, Swati Jain, Yana Zhang-Williams, Iam-Choon Khoo and Christopher J. Summers, "Tuning the optical properties of colloidal thin films using dual-frequency liquid crystal," Spring Material Research Society Meeting, San Francisco, April 17-21, 2006.
- *23. I. C. Khoo, A. Diaz and Mike Stinger, "Nonlinear Transmission of a Neat Liquid with Two-Photon and Excited State Absorption", Invited paper, ISOPL [Int. Symposium on Optical Power Limiting], June 27-29, 2006, Dingle, Ireland.
24. J. A. Bossard, L. Li, D. H. Werner, I. C. Khoo, "Infrared Liquid Crystal Tunable Frequency Selective Surfaces," Proceedings of the 2006 IEEE Antennas and Propagation Society International Symposium with USNC/URSI National Radio Science and AMEREM Meetings, Albuquerque, NM, pp 4489-4492, July 9-14, 2006.
- *25. I. C. Khoo, "Effect of excited state and population regeneration in the nonlinear transmission through a multiphoton absorbing organic liquid," Conference on Nonlinear Transmission and Multi-Photon Processes - SPIE Annual International Symposium on Optical Sciences and Technology, San Diego, CA August 13-18 2006.
26. I. Khoo, J. D. Liou, K. X. Chen, "Enhanced photorefractivity of CdSe nanorods doped nematic liquid crystals," Liquid Crystal Conference X- SPIE Annual International Symposium on Optical Sciences and Technology, San Diego, CA August 13-18 2006.

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

1. I. C. Khoo, Yana Zhang, A. Diaz, J. Ding and K. Chen, "Supra-photorefractivity of C60- and Carbon Nanotube- doped Nematic Liquid Crystals," In Liquid Crystals VII, Proceeding of SPIE International Symposium on Optical science and Technology, San Diego, California August 4-7 (2003)
2. I.C. Khoo*, A. Diaz, J. Ding, "Multiphoton and excited-state absorptions of an organic liquid for optical transmission control application," Invited Paper, in 'Nonlinear Transmission', Proceedings of SPIE International Symposium on Optical science and Technology, San Diego, California August 4-7 (2003)
3. I. C. Khoo, J. Ding and A. Diaz, "Dynamics of laser self-induced polarization conversion in nematic liquid crystals," Liquid Crystals VIII - Proceedings of SPIE Vol. 5518. Denver, CO [2004]
4. I. Khoo, Y. Z. Williams, K. X. Chen, "Nonlinear polymer dispersed liquids and liquid crystals for holographic and photonic crystal applications," Liquid Crystals VIII - Proceedings of SPIE Vol. 5518, Denver, CO. [2004]
5. A. Diaz and I. C. Khoo, "Nonlinear molecular photonics and controlled laser transmission through a nonlinear fiber array," Proceeding of SPIE European Symposium on Optics and Photonics for Defense and Security, London, Oct. 2004.
6. Yana Williams, Kan Chen, Jae Hong Park, Iam Choon Khoo, Brad Lewis, Thomas E. Mallouk, "Electro-optical and nonlinear optical properties of semiconductor nanorod doped liquid crystals," Liquid Crystal IX, SPIE Proc.Vol. 5936 [2005]

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

6

(d) Manuscripts**Number of Manuscripts:** 0.00**Number of Inventions:****Graduate Students**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
J. Ding	0.25	No
K. chen	0.25	No
Y. Z. Williams	0.25	No
A. Diaz		No
FTE Equivalent:	0.75	
Total Number:	4	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
I. C. Khoo	0.25	No
FTE Equivalent:	0.25	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent: Total Number:	

Names of Personnel receiving masters degrees

<u>NAME</u>	
Mike Stinger	No
Total Number:	1

Names of personnel receiving PHDs

<u>NAME</u>	
J. Ding	No
K. Chen	No
Y. Z. Williams	No
A. Diaz	No
Total Number:	4

Names of other research staff

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent: Total Number:	

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1. Statement of Problems Studied

Experimental and theoretical studies of recently discovered extremely nonlinear liquid crystals and novel nonlinear optical processes have been conducted in order to elucidate and completely characterize the physical origins, and to explore and demonstrate the feasibility of utilizing these unique materials in advanced multifunctional optics devices.

The multi-prong projects include: - (i) Synthesis of nonlinear nematic liquid crystals containing highly photosensitive and/or photo-conducting nano-particulates (Gold nano-wires, CdSe nanorods); (ii) Synthesis of nonlinear liquids with large two- and multi-photon absorption and excited-state absorption coefficients; (iii) Investigation and characterization of the dynamics of the mechanisms responsible for the supra-nonlinearities of these liquid crystalline systems and identification of the roles of various fields, molecular and optical parameters for optimum nonlinearity, response time and other performance characteristics (iv) Fabrication of nonlinear fiber array and tunable 2- and 3-D liquid crystalline photonic crystals. (v) Complete quantitative studies of self-action effects such as all-optical switching, optical limiting, nonlinear guided waves using visible as well as infrared lasers.

These studies have been undertaken in a cooperative integrated manner; nonlinear materials developed, and processes refined in one study have provided valuable insights and information for studies in other categories. From these concerted efforts, we have gained thorough understanding of these newly discovered supra nonlinearities of nematic liquid crystalline systems, synthesis of several highly nonlinear liquid crystalline systems, and successful feasibility demonstration of several unique and high performance holographic, modulation, mixing, limiting and adaptive optics devices. These devices are operational at low power/intensity levels over a wide spectral and temporal bandwidth, and compatible with most integrated nano-electronics and photonics components. As detailed in a recent proposal for renewal, these new findings and discoveries have opened up new avenues for future research and development possibilities, especially when coupled to nano-structures and nano-metamaterials of unique optical, electro-optical and nonlinear-optical properties.

2 Summary of Accomplishments

Liquid crystals are organic molecules that self-organize into various mesophases with different degrees of order [1]. The most commonly studied thermotropic liquid crystals manifest the smectic, nematic and cholesteric phases as a function of temperature. The most widely investigated phase is the nematic phase, in which there is directional but no positional order. The unique linear and nonlinear light scattering properties of nematic liquid crystals and their fluid nature that allow conformation to various flexible forms and shapes, and compatibility with polymeric or semiconductor materials have powered their applications in an ever widening range of optical technology as well as novel fundamental optical research.

The collective orientation of the liquid crystal molecules is described by a unit vector $\hat{n}$, the so-called director axis. They are usually assembled in thin bulk film forms, with thicknesses that could range from a few to 100's μm . The alignment of the director axis is dictated by surface anchoring agents. Nematic liquid crystals possess large and broadband birefringence and transparency [1], c.f. Fig. 1. As a result of highly developed

eutectic mixture approaches/processes, nematics with a very large operating temperature range [-40 to over 100 °C] are now readily available commercially.

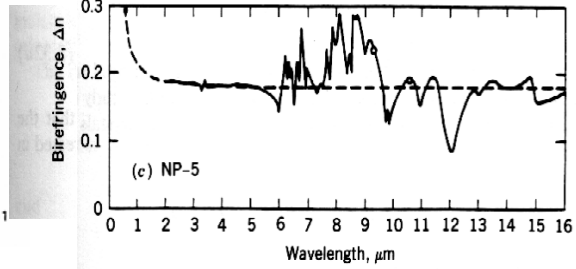
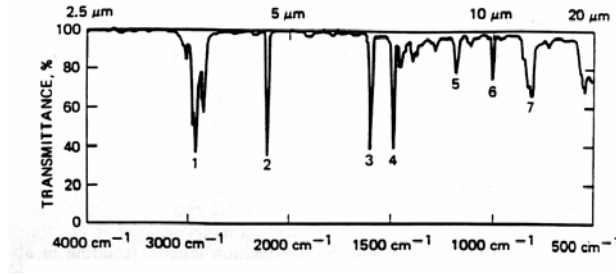


Fig. 1a Typical Transmission spectra of

Fig. 1b. Typical d birefringence of NLC

This program of studies are focused on their *extremely large optical nonlinearities* [2-9] characterized by nonlinear index changing coefficients several orders of magnitude larger than all existing materials.

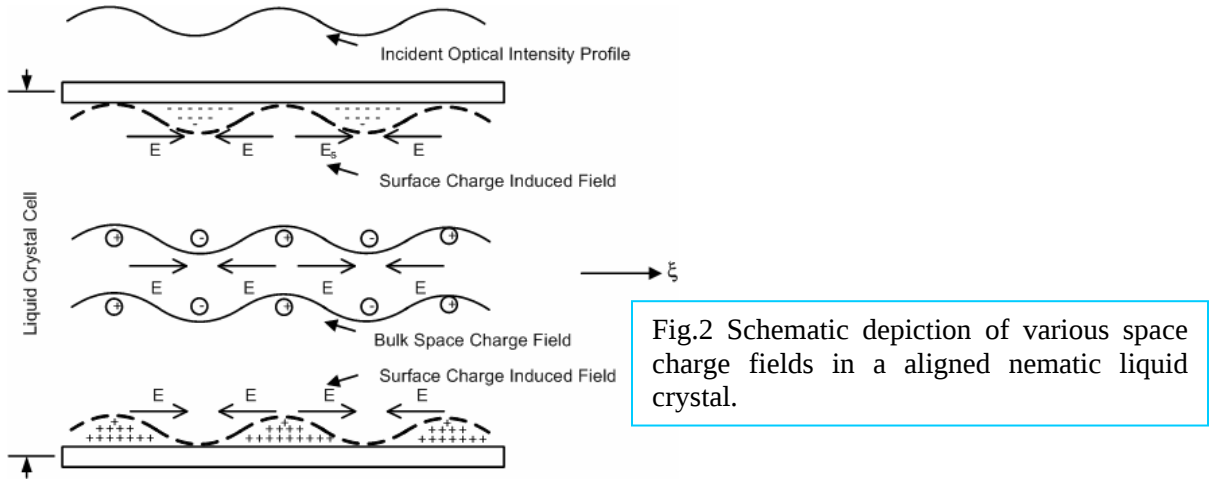
In the course of our ARO supported research program, we have made several important and noteworthy accomplishments broadly classified as follow. We have

- performed first quantitative experimental study and develop complete theoretical formalism for stimulated orientational scattering and polarization self-rotation with visible as well as infrared lasers [10, 11].
- reported large nonlinear response of gold nano-wire doped nematic liquid crystal and polymer-dispersed liquid crystals 3-D photonic crystals that are tunable and switchable, and possess multi-wavelength selective reflectivity [12].
- demonstrated the equivalence of all-optical image processing operation to feed-forward neural network signal processing model [13].
- developed the theoretical framework to describe experimental observations of two-photon mediated excited state absorption in some organic liquid and their exceptional performance [very large dynamic range and field of view] as optical limiting materials when incorporated into nonlinear fiber array [14].
- performed quantitative theoretical and experimental studies of the photorefractivity of nematic liquid crystals doped with photoconducting semiconductor nano-rods and demonstrated their extremely large nonlinear refractive index coefficients suitable for dynamic holography and image processing application [15].
- We have also observed and develop the theory for enhanced photorefractivity of liquid crystals doped with photo-conducting semiconductor [e.g. CdSe] nano-rods and develop a first quantitative theoretical model accounting for both surface and bulk space charge fields [15].
- designed nano-structures + LC overlayer for frequency selective surfaces capable of large extinction ratio optical filtering performance as well as negative index of refraction [16].
- Observed the largest electrically tunable Bragg reflection liquid crystal infiltrated 3-D nano-photonic crystals structure (inverse opals) [17].

Details can be found in the list of published refereed articles in section 4. In the following sections, we provide more details of some of exemplary findings.

2.1 Enhanced photorefractivity of CdSe nano-rod doped nematic liquid crystal

Photorefractive effects have been observed in various inorganic and organic materials. In most photorefractive materials, the mechanisms for optically induced refractive index changes are optically induced bulk space charge fields in concert with externally applied field. Most inorganic and polymeric photorefractive materials require very large external applied field [several 100's V/ μm] to activate. On the other hand, photorefractivity in nematic liquid crystals require applied field of $<1\text{V}/\mu\text{m}$], thus making them highly desirable for practical device implementation.



In liquid crystals these bulk space charge fields consist of 3 major components: one component, E_{ph} originates from photo-induced space charges and charge separation (diffusion and drift) and the other two, $E_{\Delta\sigma}$ and $E_{\Delta\epsilon}$, come from conductivity and dielectric anisotropies (Helfrich-Carr effect). For a typical optical wave mixing process involving an imparted sinusoidal optical intensity function as depicted in Fig. 2, the various fields present in a nematic liquid crystal (NLC) cell are of the form:

$$E_{ph} = E_{ph}^{(0)} \cos(q\xi) = \left[\frac{mk_B T}{2e} qv \frac{\sigma - \sigma_d}{\sigma} \right] \cos(q\xi)$$

$$E_{\Delta\sigma} = - \frac{\left[(\sigma_{\parallel} - \sigma_{\perp}) \sin \theta \cos \theta \right]}{\sigma_{\parallel} \sin^2 \theta + \sigma_{\perp} \cos^2 \theta} E_{dc}; \quad E_{\Delta\epsilon} = - \frac{\left[(\epsilon_{\parallel} - \epsilon_{\perp}) \sin \theta \cos \theta \right]}{\epsilon_{\parallel} \sin^2 \theta + \epsilon_{\perp} \cos^2 \theta} E_{dc} \quad (1)$$

where k is the elastic constant (assuming the single constant approximation), $\Delta\epsilon$ is the dc field anisotropy and $\Delta\epsilon_{op}$ is the optical dielectric anisotropy, k_B is Boltzmann constant, σ = conductivity under illumination, σ_d = dark state conductivity, and $v = (D^+ - D^-) / (D^+ + D^-)$,

where D^+ and D^- are the diffusion constants for positive and negative ions, respectively, $q = 2\pi/\Lambda$ is the grating wave vector, with Λ the grating constant.

Besides these bulk space charge fields, recent experimental have demonstrated the important role played by the optically induced surface field modulation in lowering the threshold for initiating the photorefractive effect. We have performed a quantitative analysis of the detailed torque balance equation governing the liquid crystal axis reorientation and shown that in this case, the photorefractive threshold condition becomes:

$$\left[E_{\Delta} E_{dc} \cos(\beta) + \left(\frac{\Delta \epsilon_{op}}{\Delta \epsilon} \right) E_{op}^2 \cos(2\beta) + E_S^{\text{eff}} E_{dc} \right] - E_F^2 \left[1 + \left(\frac{qd}{\pi} \right)^2 \right] > 0 \quad (2)$$

which leads to

$$E_{dc} > \left(\frac{1 + (qd/\pi)^2}{(\Delta \sigma / \sigma_{\perp} + \Delta \epsilon / \epsilon_{\perp}) \cdot \cos \beta} \right)^{\frac{1}{2}} E_F - \frac{1}{2(\Delta \sigma / \sigma_{\perp} + \Delta \epsilon / \epsilon_{\perp}) \cdot \cos \beta} E_S^{\text{eff}} \quad (3)$$

where $E_S^{\text{eff}} = \frac{E_S}{qd}$, and E_S is the surface space charge field and d the cell thickness. In terms of

the Freedericksz transition field $E_F = \frac{1}{d} \sqrt{4\pi^3 k / \Delta \epsilon}$ [or voltage $V_f = E_F d$], this gives:

$$V_{th} = \alpha V_F - \gamma E_S^{\text{eff}} d = \left(\alpha - \gamma \frac{E_S^{\text{eff}}}{E_F} \right) V_F \quad (4)$$

For 5CB, $\Delta \epsilon \sim 11$, $[\epsilon_{\parallel} \sim 16, \epsilon_{\perp} \sim 5]$, $\Delta \sigma / \sigma_{\perp} \sim 0.5$, and $[qd \sim 2\pi]$, and the internal angle $\beta = 22.5^\circ$ for a typical wave mixing geometry, equation (12) gives:

$$\begin{aligned} V_F &= 2.04V \\ V_{th} &= \alpha V_F = 1.45 \times V_F \approx 3V \end{aligned} \quad (5)$$

An important quick note on these threshold condition is the ‘smallness’ of the required threshold voltages [~ 1 volt] compared to the ~ 1000 V typically required in other inorganic or polymeric photorefractive crystals. An even more important feature is that because of the availability of many charge-producing photosensitive dopants, e.g. carbon nanotubes, CdSe nano-rods, c.f. Fig. 3, the photorefractive effect in NLC can be dramatically enhanced, c.f. Fig 4.

The observed nonlinear index coefficient n_2 of CdSe-NLC is $\sim 2.05 \times 10^{-2} \text{ cm}^2/\text{W}$, which is more than 20 times larger than that of the undoped NLC. It is comparable to C60 and porphyrin:Zn doped NLC’s and are orders of magnitude larger than all other nonlinear optical materials. We attribute this improvement to the increased charge generation capability of the photoconducting CdSe nanorods, as well as the larger dielectric and conductivity anisotropies $\Delta \epsilon$ and $\Delta \sigma$ of the doped nematic liquid crystals observed in recent experiments.

It was also observed that under prolonged illumination, the induced orientation gratings remain transient in nature, unlike their C60 or dye (e.g. methyl red) counterparts which tend

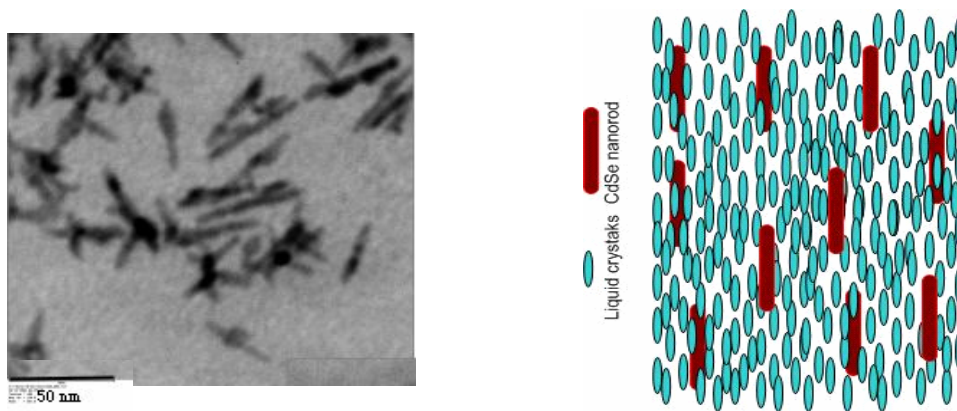


Fig.3 (left) SEM photo of CdSe nanorods. (right) Schematic depiction of planar aligned CdSe nano-rods doped nematic liquid crystal cell

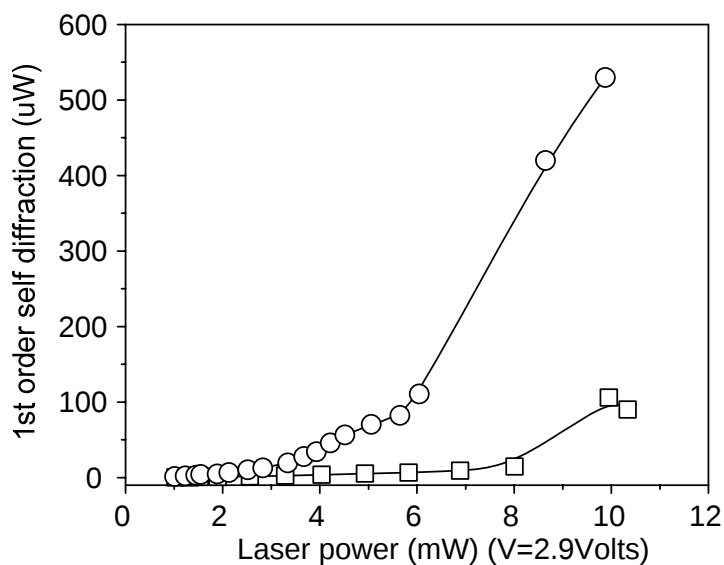


Fig.4 An example of Enhanced four wave mixing efficiency [self-diffraction] of CdSe nano-rods doped liquid crystal (circles) over undoped one [squares]

to produce unwanted persistent (or permanent) reorientation effect. This is attributed to the organic capping on the CdSe nanorods during the synthesis process; such capping inhibits adsorption of the optically excited CdSe nano-rods on the cell windows. This feature makes CdSe-doped NLCs, [and other nano-particulates to be developed in a new proposed program as well] promising candidates for real time image processing applications e.g. optical wave front conjugation, aberration correction and related adaptive optics applications.

2.2 Liquid Crystals in Nano-structures

The extraordinary optical properties of liquid crystals, when incorporated in optical structures could also lead to very dramatic modifications and creations of non-conventional optical functions and elements with very unusual properties. This is clearly demonstrated in our recent studies of LC infiltrated inverse opal photonic crystals, and frequency selective surfaces [FSS] in which large spectral tuning, electro-optical switching, frequency filtering and negative- and zero- index of refraction were realized. Fig 5a, for example, shows the first observation of large electrically tuned Bragg peak shift that can be obtained from a nematic liquid crystal infiltrated inverse opal structure, c.f. Fig. 5b.

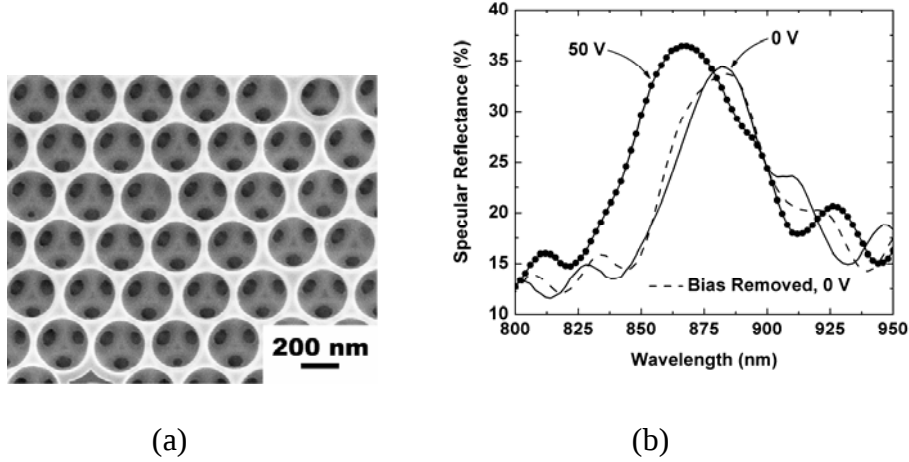


Fig. 5(a) SEM photo of a TiO₂ inverse opal 3-D photonic crystal; (b) electrically tuning of Bragg reflection peak of the inverse opal structure containing aligned nematic liquid crystals. Optical tuning is possible by means of the intensity dependent liquid crystal director axis

We have recently initiated studies of tunable optical filters and reflective/transmissive devices based on liquid-crystal-cladded frequency selective surfaces. Frequency selective surfaces [FSS's] are two-dimensional periodic arrays of metallic patches or aperture elements that possess low-pass, high-pass, band-pass, or multi-band filtering properties for incident electromagnetic waves. The planar geometry of the PFSS allows easy incorporation of a liquid crystal overlayer. Fig. 6 shows the tunable filter action of a liquid-crystal-cladded FSS designed for the near infrared region; the stop-band in the transmission response can be shifted from 92 THz [$\sim 3.26 \mu\text{m}$] to 83 THz [$3.64 \mu\text{m}$], i.e. a tuning range of 380 nm for a liquid crystal birefringence Δn of 0.6. Accordingly, since the birefringence of nematic liquid crystals span the **entire visible to far infrared spectrum**, LC-PFSS will allow us to design and fabricate very broadband high-extinction-ratio **tunable** filters/switches for polarized light.

As reported in [16], we have designed a structure that is capable of exhibiting negative index of refraction in the 90 -110 THz region [optical wavelength $\sim 3.3 \mu\text{m}$ - $2.7 \mu\text{m}$]. By incorporating liquid crystal as an overlayer onto these NIM structures, it is clearly feasible to design LC-NIM at any desired frequency in the entire visible-infrared regime. Since the dielectric constant of the liquid crystal can be electrically modulated,

these NIMs possess the interesting and potentially very useful property that they can be switched between the positive and the negative index states.

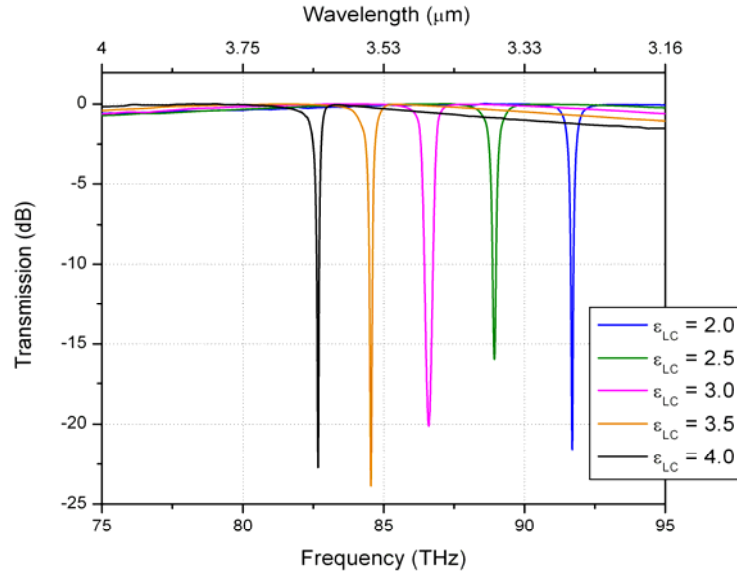


Fig.6 Transmission spectrum of a nematic liquid crystal (NLC) clad FSS structure for the range of dielectric constant of NLC showing very widely tunable optical filtering capability of the structure..

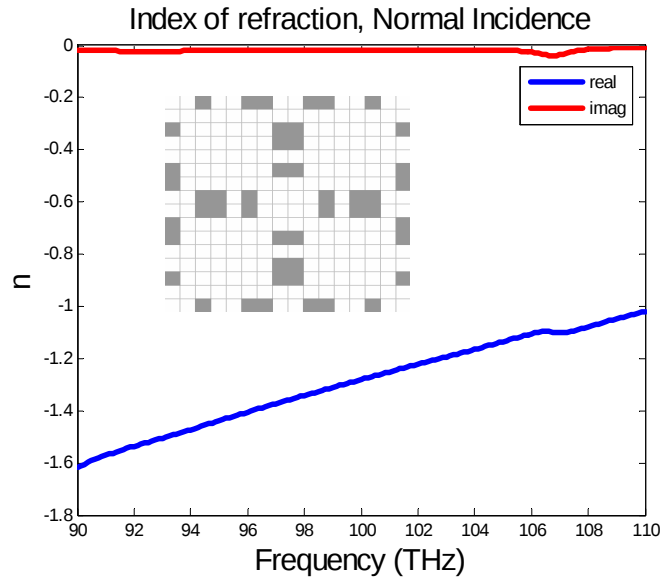


Fig.7 Real and Imaginary parts of the refractive index of an all-dielectric FSS. Note: the convention used [plane wave represented as $\exp(-ikz)$] is such that the imaginary part is negative in a lossy medium.

Besides periodic structures such as FSS's and 3-D photonic crystals which require complex nano-fabrication techniques, we have recently also studied a configuration involving 'free' propagation of polarized light through aligned nematic liquid crystal cells in which nano-spheres are dispersed, c.f. Fig 8a. The combination of the permittivities at the appropriate resonances, in conjunction with the field induced permittivity change in the LC host that give rise to the effective refractive index of nano-sphere dispersed liquid crystals (NDLC) that can vary from negative-zero-positive values.

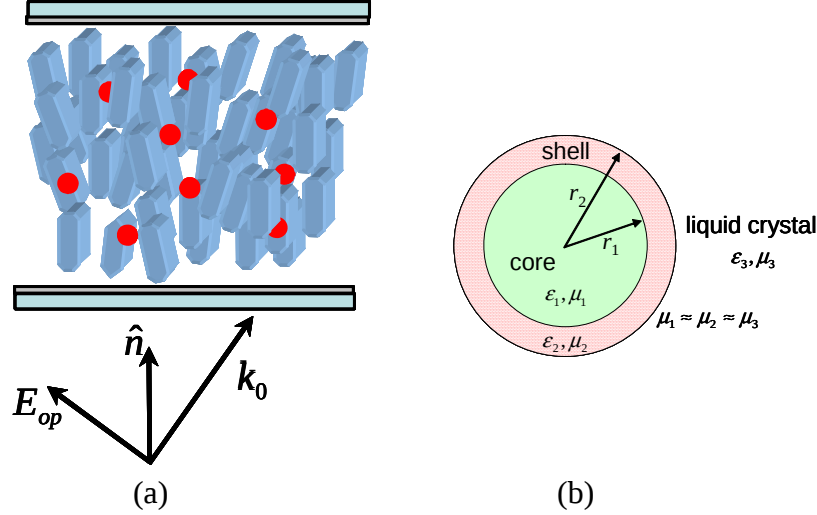


Fig. 8(a) Schematic of polarized light incident as an extraordinary wave on an aligned nematic liquid crystal containing nano coated spheres. (b) Dimensions and composition of the core-shell nano-spheres.

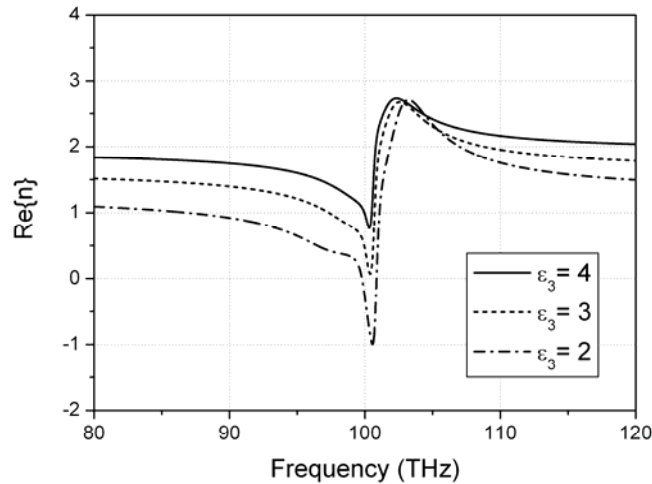


Fig. 9. Calculated real part of the refractive index of nano coated-spheres dispersed nematic liquid crystal film as the director axis orientation is realigned [electrically or optically].

2.3 Ultrafast electronic optical nonlinearities of isotropic liquid crystals for optical limiting application

As organic materials, liquid crystals also possess ultrafast electronic optical nonlinearities that rank among the largest of all known materials. What is even more important for optical limiting application is that the molecular constituents making up liquid crystals in general possess, *simultaneously*, large two-photon absorption and excited state absorption capabilities. In the course of the ARO program, we discovered an isotropic liquid crystal, L34 that possess all the characteristics of such an ideal non-linearly absorbing optical limiter. Fig. 10 shows the molecular structure and absorption spectrum of L34. It is transparent in the entire visible regime.

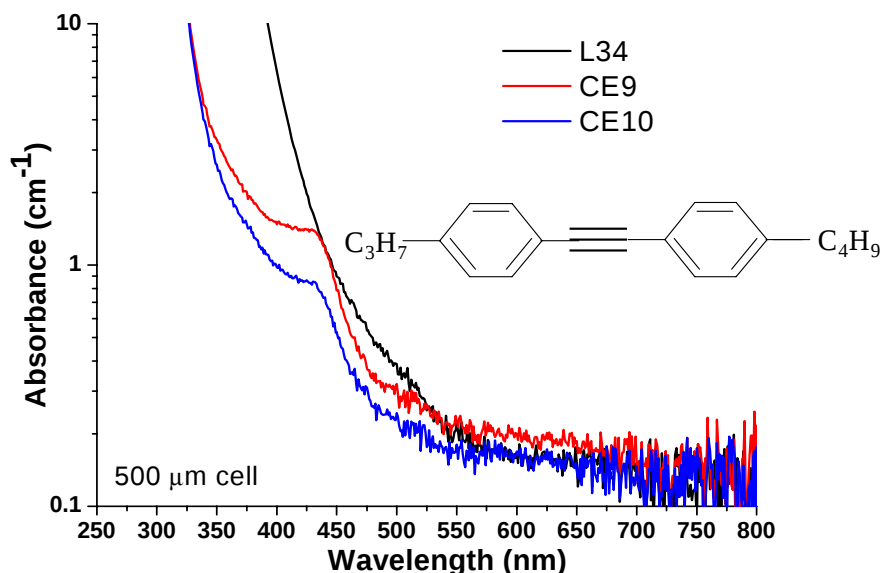


Fig.10 Molecular structure of L34 and its linear absorption spectrum in the visible region. Also shown are the absorption spectra of two variants of L34.

Recent Femto- and pico-second nonlinear transmission (z-scan) studies [18]⁴¹ of L34 revealed that the intrinsic TPA coefficient $\beta \sim 4.7$ cm/GW whereas nanosecond measurements showed that β is intensity dependent, increasing from the intrinsic value to much higher value of over 25 cm/GW, which ranks among the largest of all neat liquids, and will enable very low threshold optical limiting performance.

In conclusion, the program has resulted in the development of several supra-nonlinear nematic liquid crystalline materials, and nonlinear liquids with fast and efficient nonlinear absorption or index changing properties, as well as detailed quantitative formulation and illustration of several novel nonlinear optical phenomena and processes. These materials and detailed understanding of the nonlinear processes will enable a host of multi-functional optical devices that operate with very low power thresholds, and are applicable over broad spectral and temporal bandwidths. Examples are all-optical switches and beam/image processor that operate under μ W optical power,

large dynamic range nonlinear fiber array for optical limiting application against agile frequency pulsed lasers, and electro-optically or self-action optical switches and beam steering devices.

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2. I.C. Khoo and A. Diaz, "Nonlinear dynamics in laser polarization conversion by stimulated scattering in nematic liquid crystal films," *Phys. Rev. E* 68 pp 042701-1 to -4 [2003]
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 - *13. I. C. Khoo, "Liquid crystals-Photonic crystals" Invited paper. Annual Meeting of the IEEE-Lasers and Electro Optics Society, Nov. 8, 2004, Puerto Rico
 - 14. I. C. Khoo, "All-Optical Self-Polarization Switching via two-wave mixing in nematic Liquid Crystals – from visible to infrared", Annual Meeting of the IEEE-Lasers and Electro Optics Society, Nov. 11, 2004, Puerto Rico.
 - *15. I. C. Khoo, "Holographic fabrication of polymer dispersed liquid crystal photonic crystals," Invited paper, Materials Research Society Fall Meeting, Boston, Dec. 2, 2004
 - *16. I. C. Khoo, "Liquid Crystals Ultra Broadband All-Optical Devices", 8th Int. Conference on Frontiers of Polymers and Advanced Materials, Cancun, Mexico April 23-28, 2005.
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 - *18. I. C. Khoo, "Electro- and Nonlinear –optical Properties of Liquid Crystals," 7th Int. Topical Meeting on Novel Optical Materials and Applications," Cetraro, Italy. May 29 – June 4, 2005.
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- *25. I. C. Khoo and A. Diaz, "Self-action cw-pulsed lasers and glares attenuators," Invited paper, IEEE-Lasers and Electro-Optics Society Annual Meeting, Sydney, Australia [10/2005].
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- *27 I. C. Khoo, A. Diaz and Mike Stinger, "Nonlinear Transmission of a Neat Liquid with Two-Photon and Excited State Absorption", Invited paper, ISOPL [Int. Symposium on Optical Power Limiting," June 27-29, 2006, Dingle, Ireland.
28. J. A. Bossard, L. Li, D. H. Werner, I. C. Khoo, "Infrared Liquid Crystal Tunable Frequency Selective Surfaces," Proceedings of the 2006 *IEEE Antennas and Propagation Society International Symposium with USNC/URSI National Radio Science and AMEREM Meetings*, Albuquerque, NM, pp 4489-4492, July 9-14, 2006.
- *29. I. C. Khoo, "Effect of excited state and population regeneration in the nonlinear transmission through a multiphoton absorbing organic liquid," Conference on *Nonlinear Transmission and Multi-Photon Processes* - SPIE Annual *International Symposium on Optical Sciences and Technology*, San Diego, CA August 13-18 2006.
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31. I. Khoo, J. D. Liou, K. X. Chen, “Enhanced photorefractivity of CdSe nanorods doped nematic liquid crystals,” *Liquid Crystal Conference X- SPIE Annual International Symposium on Optical Sciences and Technology*, San Diego, CA August 13-18 2006.

6. Invention Disclosure.

- I. C. Khoo and A. Lei “Extremely low freezing point non-volatile nonlinear optical liquids for eye/sensor protection against intense laser pulses” # 2003-2854 Filed on 11/26/2003.
- I. C. Khoo, Z. Liu and S. Yin, “Electronically tunable and highly nonlinear photonic crystal fiber.” PSU Invention Disclosure #2003-2843 Filed Oct. 8, 2003 [individual initiative]

7. Technology Transfer

- Consultant to a Army Research Office SBIR Phase II program on nano- dispersed supra-nonlinear liquid crystals for spatial light modulation awarded to BEAM Engineering Corporation, Winter Park, Florida. Completed 8/2005.
- Consultant to a Air Force Research Laboratory [WPAFB] SBIR Phase II program on sensor protection and optical limiting awarded to BEAM Engineering Corporation, Winter Park, Florida. Project completed in 9/2005.
- Principal investigator in a Cooperative Agreement with the Navy Air Development Center, Patuxent River, Maryland to develop a helmet-mount eye protection goggle in collaboration with SRI International, Menlo Park, CA. Project completed in 7/2006.

8. Participating Scientific Personnel

Principal Investigator: Prof. I. C. Khoo
Post-doc: Andres Diaz and J. H. Park
Graduate students: J. Ding, Y. Zhang, K. Chen, J. Liou.

9. Graduate Theses

Ph. D. Theses by: Jiangwu Ding, Yana Zhang, Kan Chen, Andres Diaz
MS thesis: Mike Stinger