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In nematic liquid crystal films doped with methyl-red dye or azobenzene liquid crystal, we have observed extremely efficient optically induced director axis reorientation effects, and refractive index change coefficients >> 1 cm^2/W. The basic mechanisms involved are determined to be optically induced space charge fields and flow reorientation, order parameter modification and molecular torques by the photo-excited dopant molecules. These supra-optical nonlinearities (SONs) enable nonlinear image conversion, optical limiting and holographic grating formation with μW power lasers.

Keywords: liquid crystal; supra-optical nonlinearity; methyl red; azobenzene liquid crystal; refractive index change; image conversion; optical limiting

INTRODUCTION

Nematic liquid crystals posses very broadband birefringence and transparency [1,2], spanning the visible to infrared spectrum [0.4 microns - 12 microns]. Because of their easy susceptibility to optical fields, they are also highly nonlinear optical materials. By far laser induced director axis reorientation gives the largest refractive index change Δn for a given optical intensity I, the so called nonlinear index coefficient $n_2 = \Delta n/I$. Recent studies have revealed several interesting effects, including 'photorefractive-like' refractive index change in dye-or fullerene C60- doped nematic liquid crystals films [3-5]. Observation of perhaps the largest nonlinear index change mechanism in aligned methyl-red doped nematic liquid crystal film [8] has also been reported. More recently, we have observed an equally large nonlinear index coefficient in azobenene liquid crystals (ALC) doped nematic liquid crystal. In planar aligned films, we found that holographic index gratings can be efficiently generated by co- as well as cross-polarized writing beams.

OPTICALLY INDUCED REFRACTIVE INDEX CHANGE.

Consider a standard grating diffraction set up in which two polarized coherent writing beams are overlapped on a nematic film, which is probed by a polarized reading beam as shown in **Fig. 1a.** If the writing beams are co-polarized, they impart an optical

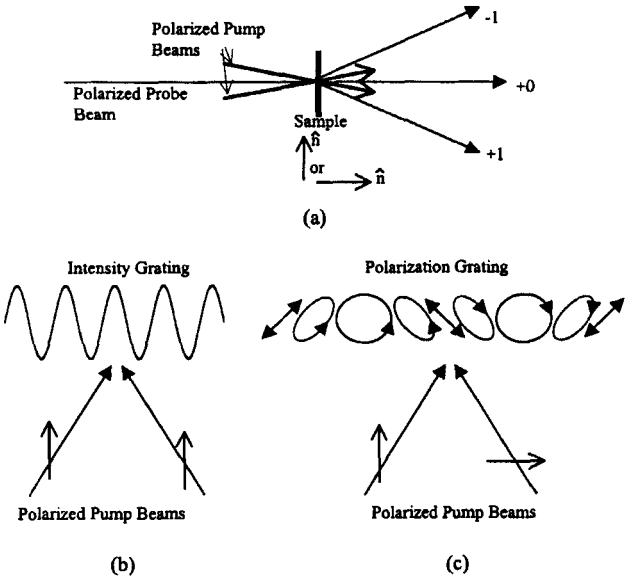

Fig. 1. a.) Schematic depiction of grating formation and probe diffraction from the index grating induced in a nematic film. **b.)** Optical Intensity grating formed by interfering two coherent co-polarized writing beams. **c.)** Optical polarization grating formed by interfering two coherent crossed-polarized writing beams

intensity grating onto the nematic film $I = I_O (1 + \cos qx)$, where q is the grating wave vector, c.f. **fig 1b**. On the other hand, if the writing beams are orthogonally polarized, the resulting intensity is uniform over the illuminated area, and instead, a polarization grating is created, c.f. **Fig. 1c**. The polarization state of the light in the overlapped region varies from circular, through elliptical to linear in accordance to the phase difference between the two coherent writing beams. Using a linearly polarized probe beam, this set up allows one to distinguish various intensity and reorientation dependent effects.

The writing beams could change the refractive index (as sensed by the probe) of the nematic film through a variety of processes. These include:

1. Reorientation by dielectric optical torque $-M_{op}$
2. Reorientation by photo-excited dopant molecular torque - M_{dopant}
3. Quadratic Photorefractive effects associated with photo-induced space charge field - E_{sc}
4. Flow-reorientation effect due to ionic flows caused by space charge fields and/or applied field. The shear torque M flow = - [gradient V] $f(\theta, \alpha, ...)$
5. Order parameter change ΔS caused by trans-cis isomerization of the photo-excited dopant molecules

In purely dielectric non-absorbing nematic liquid crystal, director axis reorientation is caused by the torque M_{op} originating from the dipolar interaction between the dielectric anisotropy $\Delta\varepsilon$ and the optical electric field. This effect was first discovered almost two decades ago, and was termed Giant Optical Nonlinearity (GON) then. The observed n_2 values in planar or homeotropic samples are on the order of 10^{-4} cm^2/Watt. If the nematic liquid crystals are doped with some dyes capable of excited-state conformational changes, studies[7,8] have shown that the excited dye molecules could exert an intermolecular torque M_m of much larger magnitude than M_{op}, resulting in a nonlinear index coefficients n_2 of 10^{-3} cm^2/W.

PHOTOREFRACTIVE EFFECT - Space Charge fields, flows reorientation

Photorefractive-like nonlinearity occurs in nematic liquid crystals lightly doped with charge producing agents [3-5] such as Fullerene C_{60}. For a 25 μm thick homeotropically aligned film under a dc bias voltage of 1.5V, the observed nonlinear coefficient n_2 is in the range of 10^{-3} - 10^{-2} cm^2/W. At the time of its observation, it was ranked as the largest among all known nonlinear optical materials. The basic mechanisms are illustrated in **Fig. 2**. An incident optical field excites the doped liquid crystals to form charge transfer complexes (CTC). The CTCs subsequently dissociate[5,9] and create dc space charge fields through ionic diffusion, migration and other electrodynamical processes. The space charge fields Esc, in combination with an applied dc field Edc, cause director axis reorientation via the quadratic field effect, i.e., the reorientation is proportional to EE.

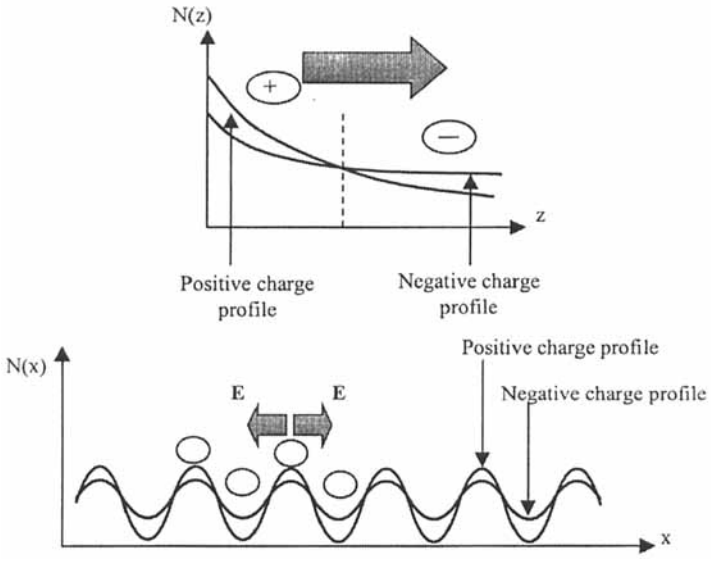

Fig. 2. Schematic depiction of photo-induced space charge field formation in nematic liquid crystal illuminated by sinusoidal laser intensity. Upper diagram: along the beam propagation direction. Lower diagram: along the grating direction.

For an incident optical intensity grating function of the form $I_{op} = I_o(1+m\cos q\xi)$, the photorefractive-like space charge field it is given by:

$$E_{ph} = q\nu m k_b T \frac{\sigma - \sigma_d}{2e\sigma} \cos\left(qx - \frac{\pi}{2}\right) \qquad (1)$$

where k_b = Boltzmann constant, σ = illuminated conductivity, σ_d = dark state conductivity, $\nu = (D_+ - D_-)/(D_+ + D_-)$, 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 the grating wave vector.

Note that there is a $\pi/2$ **phase shift** between the incident optical intensity function and the photorefractive space charge field (and the resultant refractive index change). This phase shift is responsible for two-beam coupling effect in which there is unidirectional flow of energy from one beam to the other[3,10].

Two other sources of space charge fields arise from the conductivity and dielectric anisotropies of the nematic under the action of the dc bias field E_{dc}[3, 11]:

$$E_{\Delta\sigma} = E_{dc}\frac{(\sigma_{\parallel} - \sigma_{\perp})\sin\theta\cos\theta}{\sigma_{\parallel}\sin 2\theta + \sigma_{\perp}\cos 2\theta} \tag{2a}$$

$$E_{\Delta\varepsilon} = E_{dc}\frac{(\varepsilon_{\parallel} - \varepsilon_{\perp})\sin\theta\cos\theta}{\varepsilon_{\parallel}\sin 2\theta + \varepsilon_{\perp}\cos 2\theta} \tag{2b}$$

where $(\sigma_{//} - \sigma_{\perp})$ is the **conductivity anisotropy** and $(\varepsilon_{//} - \varepsilon_{\perp})$ the **dielectric anisotropy** of the liquid crystal, and θ is the reorientation angle.

In addition to this field-induced director axis reorientation effect, the shear torque associated with the flow of the charged ions and complexes under the dc space charge fields and the applied dc field also contributes to enhancing the reorientation initiated by the space charge field. Under continuous illumination, we observed that these flows induced reorientation is the principal mechanism leading to a very large director axis reorientation in the steady state.

In the transient domain, when the reorientation angle is small, flows and the Carr Helfrich field effects may be ignored, the reorientation angle θ_o induced in conjunction with an applied dc field for the configuration as depicted in **Fig. 2c** is given by[3,4]:

$$\theta_0 = \frac{\Delta\varepsilon E_z E_{sc}\cos(\beta)}{K\left[\frac{\pi^2}{d^2} + q^2\right]} \tag{3}$$

Since E_{sc} varies as q, therefore $\theta_o \sim q/[\pi^2/d^2 + q^2]$ and it reaches a maximum at a grating spacing $\Lambda_{max} \sim 2d$. On the other hand, in the long time case where the reorientation angle is large, and flows and Carr-Helfrich effects are appreciable, the reorientation angle accounting for all the space charge fields, flows induced reorientation ....etc. is given by [4]:

$$\theta = \frac{\Delta\varepsilon E_z E_{sc}(0)\cos(\beta)}{K\left[\frac{\pi^2}{d^2}+q^2\right]+\frac{\Delta\varepsilon E_z^2}{4\pi\left[1+(\Delta\varepsilon/\varepsilon_1+\Delta\sigma/\sigma_2)\cos(\beta)+\alpha_3\Delta\sigma\varepsilon_2/(\eta_2\sigma_1\Delta\varepsilon)\right]}} \quad (4)$$

Using typical values [1,3] for K, $\Delta\varepsilon$, ε_1, $\Delta\sigma$, σ_2, α_3, η_2, etc., the second term in the denominator is estimated to be on the order of $K\pi^2/d^2$. In other words, we have

$$\theta_0 \sim \frac{q}{K\left[\frac{2\pi^2}{d^2}+q^2\right]} \quad (5)$$

Notice that in this case, corresponding to long term illumination of the sample by the writing beams, the maximum orientation will occur if the grating spacing $\Lambda_{max} \sim 0.717$ d. Both relationships (3) and (5) have been verified in our studies of transient and storage gratings in C40 doped nematic liquid crystal films [3].

SUPRA-NONLINEARITY IN METHYL-RED DOPED NEMATIC LIQUID CRYSTALS

Recent studies[6,12-14] in methyl-red doped nematic liquid crystal [MRNLC] films have ushered in the era of supra-nonlinearity. Without the need for a dc bias field, a nonlinear index coefficients $n_2 \sim 10$ cm^2/Watt [$\chi^{(3)} \sim 300$ esu], that is 10^4 times larger than the previous record, was obtained in standard grating diffraction experiments, c.f. Fig. 1. The effect is attributed to director axis reorientation caused by the photo-induced space charge fields in these films. The high photo-charge production efficiency in MRNLC is manifested in a sizable dc photo-voltages detected across the ITO coated windows, c.f. Fig. 3. Larger anisotropies $\Delta\varepsilon$, σ-σ_d, ν', $\Delta\sigma_{dc}$, $\Delta\varepsilon_{dc}$..etc, c.f. equations 1 -2, may also play a role.

A measure of the internal dc apace charge field strength may be obtained from the dependence of the probe diffraction on an applied dc voltage across the 25 μm thick nematic film. From Fig. 4, note that a dc voltage on either direction of ~ 1Volt, i.e., a field strength of ~ 400 Volt/cm, completely diminishes the effect, implying that the internal field along the z-direction is on this order. The field strength between the grating maxima and minima is even larger, since the intensity contrast is much higher between the intensity maxima and minima, than between the two cell windows. In other word, for comparable grating spacing and thickness, the field along the grating wave vector is the dominant one.

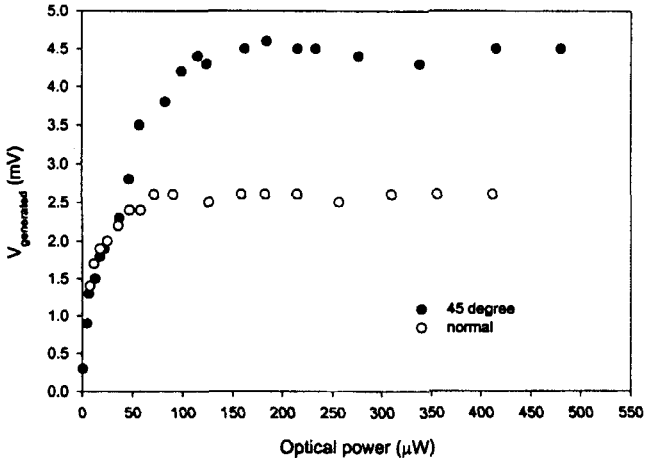

Fig. 3. The dependence of photo-voltage from a 5% Methyl-red doped homeotropic aligned 5CB film on the optical power for normally and obliquely incident light. Beam diameter: 9 mm. Sample thickness: 20 μm.

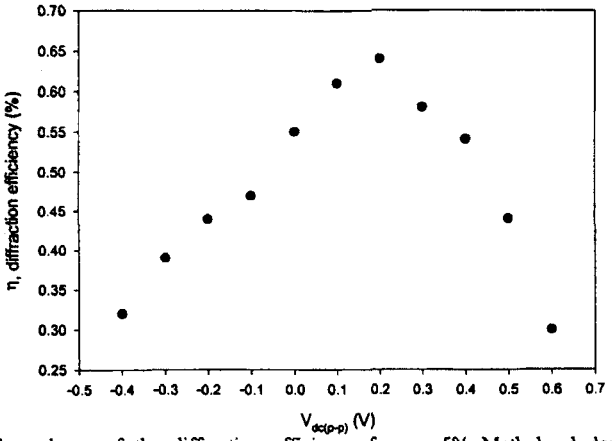

Fig. 4. The dependence of the diffraction efficiency from a 5% Methyl-red doped homeotropic aligned 5CB film on the optical power for obliquely incident extra-ordinary writing and probing beams. Sample thickness: 20 μm.

That the effect is due to director axis reorientation is confirmed by polarization dependence of the diffraction, temperature dependence, and the relaxation times [6]. In general, the reorientation is along the direction of the grating wave vector, and is independent of the writing beam polarization for normally incident writing beams [β=0]. For example, consider the geometry shown in Fig. 3. If the polarization of pump beams is o- or e-, and grating wave vector is along x, the director axis orientation is along x, and only e-polarized probe will yield diffraction. Temperature dependence of the photovoltage and diffraction also confirm that under the low writing optical intensity used, there is negligible contribution from thermal effect [14].

There is, however, a notable difference between MRNLC and photorefractive nematic liquid crystals. While they share the commonality of photo-charge and -voltage production, there is, however, no phase shift between the optical intensity function and the resultant spatial index change created in MRNLC, in contrast to the orientational photorefractive effect. Many attempts to observe beam coupling effects in MRNLC have so far yielded null effect. This and the fact that the reorientation is always along the grating wave vector have led us to conclude that flow-reorientation effect also plays a significant role in this case, c.f. discussion following equation (5). The space charge fields causes flows of the charge carrying dopant and liquid crystal molecules, creating shear torque that in turns reorients the LC director axis. In fact, we have observed similar dependence of the diffraction efficiency on the grating constant as the orientational photorefractive effect; the diffraction efficiency is maximal when the grating constant is twice the sample thickness[3,6]. In a later publication, details of the calculation and analysis of the experimental results will be given.

Contribution from Trans-Cis Isomerization, order parameter change and molecular torques ?

Since methyl-red are azo-dyes, they will undergo trans --> cis isomerization, c.f. **Fig. 5**. It is well known that the cis species are structurally incongruous with the nematic order and therefore induce a local disorder at the light intensity maxima [15,16]. The resulting director axis reorientation is therefore random, and thus both polarization state of the probe laser will yield diffraction, as the following section on similar grating diffraction experiment in azobenzene liquid crystal (AZOLC) - doped nematic films will show. As pointed out in the preceding section, in methyl-red doped nematic films, we observed that the reorientation occur only along the grating wave vector direction in the low optical intensity regime [1 mW/ cm^2], and therefore such 'disorder' mechanism is not likely to be the involved. We did observed that at higher optical intensity, (10's of mW/cm^2), the trans-cis isomerization mechanisms do manifest in a manner similar to those reported in reference [15]. In this case, both polarization state of the probe beam will yield diffraction.

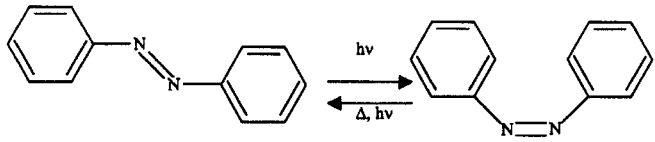

Fig. 5. Trans-cis isomerization of AZO –molecules upon optical illumination.

One possibility that could occur even at low light intensity is the molecular torque exerted by the photo-excited dye molecules on the NLC. That this mechanism does play a role is clearly demonstrated in experiments using an input polarization grating on planar sample. In the case of planar sample with one hard anchoring surface and one soft anchoring surface, it is found that the methyl-red surface alignment on the soft anchoring surface could be modulated by such polarization grating, in which the optical intensity is spatially uniform. Because of the presence also of a z-component dc space charge field, it is observed that prolonged illumination of the sample lead to permanent surface realignment, i.e., a permanent grating, as the excited dyes are adsorbed on the window surface [12,13].

AZOBENZENE LIQUID CRYSTAL DOPED NEMATIC NONLINEARITIES.

We have conducted a series of experiments using an azobenzene LC, 4.4 butylmethoxy azobenzene (BMAB), as a dopant, and in the process, 'discovered' another supra-nonlinear nematic liquid crystalline material. AZOLC is known to exhibit very well defined Trans-Cis isomerization process [16] under photo-illumination. The absorption spectrum of BMAB is shown in **Fig. 6**. The absorption maximum near 350 nm corresponds to a $\pi - \pi^*$ transition whereas the absorption peak near 450 nm is due to a n $-\pi^*$ transition. Under illumination by a linearly polarized laser [λ = 488 nm], the AZOLC molecule will tend to realign with its transition moment along the optical electric field in order to minimize the energy.

When BMAB is dissolved in the nematic host, and the resultant sample is exposed to the polarized writing beams, we observe that there are two main mechanisms for creating director axis reorientation and refractive index change. One is due to Trans – Cis isomerization. The Cis species are incongruous to the nematic order, and thus causes order parameter change, and therefore refractive index modification. We found that at high illumination intensity 10's of mW/cm^2], sufficient Cis species will be created to cause nematic - isotropic phase transition. The other index changing mechanism is the tendency of the Trans molecules to align with its transition moment perpendicular to the optical polarization [i.e., the long axis parallel to incident light polarization] in order to minimize the interaction (absorption) energies. This dye molecular realignment causes the NLC director axis to follow.

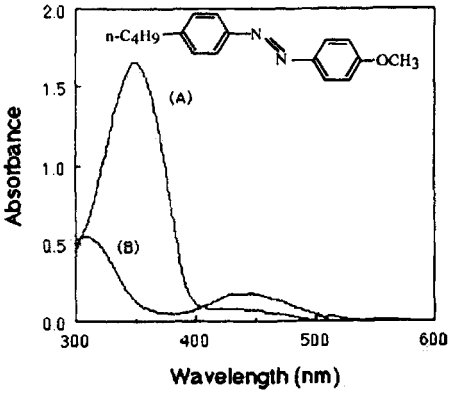

Fig. 6. Absorption spectra of Trans and Cis species of the azobenzene liquid crystals in isotropic phase. Insert is the molecular structure.

Using Fig. 1 set up, we have studied these director axis reorientation effects and index changes. The following is a summary of the observation and analysis. Planar samples of AZOLC doped 5CB or E& nematic liquid crystals are made with standard rubbed polymer-coated cell windows. For the mixture to remain in the nematic phase, the maximum concentration by weight of AZOLC is about 18 % in 5CB, and 10 % in E7. The film thickness used is 10 microns. These dye-doped samples exhibit dichroism. Typically, the transmission for light polarized along the director axis is about three times less than for light polarized perpendicular to the director axis.

Optical Intensity Grating. In the case of an imparted intensity grating, we found that both order parameter change and molecular realignment effect contribute. Probing the sample with beam polarized as e- or o-wave yielded diffraction. In general, if the probe polarization is parallel to the initial director axis direction, it will experience larger refractive index modification and yield a higher diffraction efficiency. Because of diffusion of the Cis species from the intensity maxima and to the minima, the induced grating tends to be degraded. A typical dynamics of the diffraction is shown in Fig. 7a. The diffraction rises to a peak and then falls to a much smaller steady state final value.

Optical Polarization Grating. The diffusion and grating degradation problem is eliminated if an optical polarization grating is used. In this case, uniform intensity means that the order parameter change is also uniform, i.e., no grating arise from this mechanism. Therefore, the director axis reorientation is due to the molecular torque exerted by the illuminated trans AZOLC molecules, which tend to align parallel to the

optical field polarization vector as we explained earlier. These molecular torques reorient the nematic director axis. Since there is no intensity grating in this case, grating degradation due to diffusion is not present, and the diffraction signal usually could build up to a large steady state value, as shown in fig. 7b. We do note that at high intensities (10's of m/cm2), the polarization grating will be wiped out because the whole sample turns isotropic due to sufficient concentration build up of the cis species.

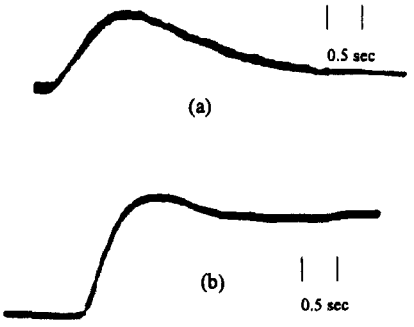

Fig.7. **a.)** Oscilloscope trace showing the dynamical behavior of the first order probe diffraction from the AZOLC doped nematic film induced by an optical intensity grating. Sample thickness: 10 μm. Probe polarization parallel to initial director axis orientation. **b.)** Oscilloscope trace showing the dynamical behavior of the first order probe diffraction from the AZOLC doped nematic [5CB] film induced by an optical polarization grating.

We close this section with an estimate of the nonlinear index coefficient of AZOLC doped NLC film. We observed that the best diffraction efficiency is obtained if the writing beam is normally incident on the planar sample, with the NLC director axis at 45 degrees to the x-axis. In this case, we get a diffraction efficiency of $\eta = 1$ % for a writing beam intensity of 1 mW/cm^2 from a 10 micron thick sample. Since $\eta = (\pi \Delta n d/l)^2$, we get $\Delta n = 2 \times 10^{-3}$, and therefore a nonlinear index coefficient $n_2 = 2$ cm^2/Watt. With some optimization, much higher values of n_2 could be obtained.

Nonlinearity of Nematic Liquid Crystals

The nonlinear coefficient n_2 of nematic liquid crystals and other well-known nonlinear optical materials [17-19] are listed in Table 1.

Table 1. Refractive Index Coefficients of Nonlinear Optical Materials
Material and Nonlinearities order of magnitude n_2 (cm^2/W)

Nematic Liquid Crystals	
Purely optically induced [ref.1]	10^{-4}
Excited dye molecular torque [ref.13]	10^{-3}
Photorefractive -C60 doped [ref.14]	10^{-3}
methyl-red doped [ref.3]	**10**
AzoLC-doped	**2**
Isotropic Liquid Crystals [ref.1]	10^{-11}
GaAs bulk [ref.17]	10^{-5}
GaAs MQW [ref.18]	10^{-3}
Photorefractive crystals [ref.19]	10^{-4}
CS_2 [ref.1]	10^{-13}

These supra nonlinearities have been utilized for image processing with $\mu W/cm^2$ intensities, and optical limiting application with high dynamic range [14], following our previous observation of self-defocusing and optical limiting effect with nanowatt power laser [20]. An attractive feature of such imaging conversion process using MRNLC film is that visible image can be reconstructed with an input beam intensity as low as 70 $\mu Watt/cm^2$. This sensitivity is among the best of all known optical processing materials, and rivals those obtainable from commercial [and much more expensive] liquid crystal spatial light modulator LCSLM[21,22] used in adaptive optics systems.

CONCLUSION

In conclusion, nematic liquid crystals in its various pure and doped forms possess many interesting nonlinear optical responses. In methyl-red or azobenzene liquid crystal doped nematic films, we have observed supra-optical nonlinearity characterized with index coefficients that are orders of magnitude larger than all known nonlinear optical materials.

ACKNOWLEDGEMENT

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