Laser-induced cumulative birefringence effects in nematic liquid crystal: 10-Hz pulse repetition rate

Svetlana G. Lukishova and Robert W. Boyd
The Institute of Optics, University of Rochester, Rochester, NY 14627-0186
phone: 716/275-5030; FAX: 716/244-4936; E-mail:lukishov@optics.rochester.edu

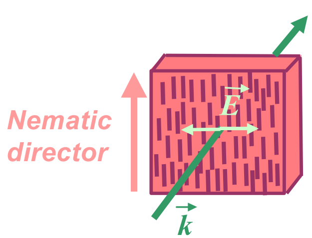
Kenneth L. Marshall
Laboratory for Laser Energetics, University of Rochester, 250 East River Road, Rochester, NY 14623-1299

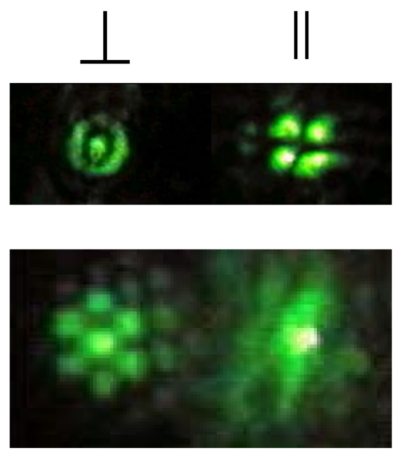
QELS'2002, Long Beach, CA

Abstract: Laser-induced cumulative birefringence effects in nematic liquid crystals: 532-nm, 15 ns, 10-Hz pulse repetition rate

Birefringence effects induced by repetitive nanosecond laser radiation in highly absorbing planar-nematic layers at incident intensities I ~ 0.001 - 0.05 GW/cm^2 with polarization perpendicular to a nematic director manifest themselves in:

(1) the appearance of a polarization component parallel to nematic director;
(2) two different modes of feedback-free spatial pattern formation with different patterns for parallel and perpendicular polarization





Cumulative nature of these effects manifests itself in a buildup time of several seconds to minutes

Motivation and objective

- *Cumulative* effects are seldom studied for the *l0-Hz* pulse repetition rate that is common in nanosecond optical power limiting applications and z-scan measurements. However, recently they were reported [1, 2] for liquid-crystal thermal nonlinearity.

- The present paper describes another manifestation of cumulative effects in heating of liquid crystals by <u>low-repetition-rate</u> nanosecond radiation. It appears as induced-birefringence effects and spatial pattern formation.

1. C. Umeton, G. Cipparrone and F. Simoni, "Power limiting and optical switching with nematic liquid-crystal films", *Opt. Quant. Electr*., 18, 312 (1986).

2. S.G. Lukishova, "Nonlinear optical response of cyanobiphenyl liquid crystals to high-power, nanosecond laser radiation", *J. Nonl. Opt. Phys.& Mater*., 9, no 3, 365-411 (2000).

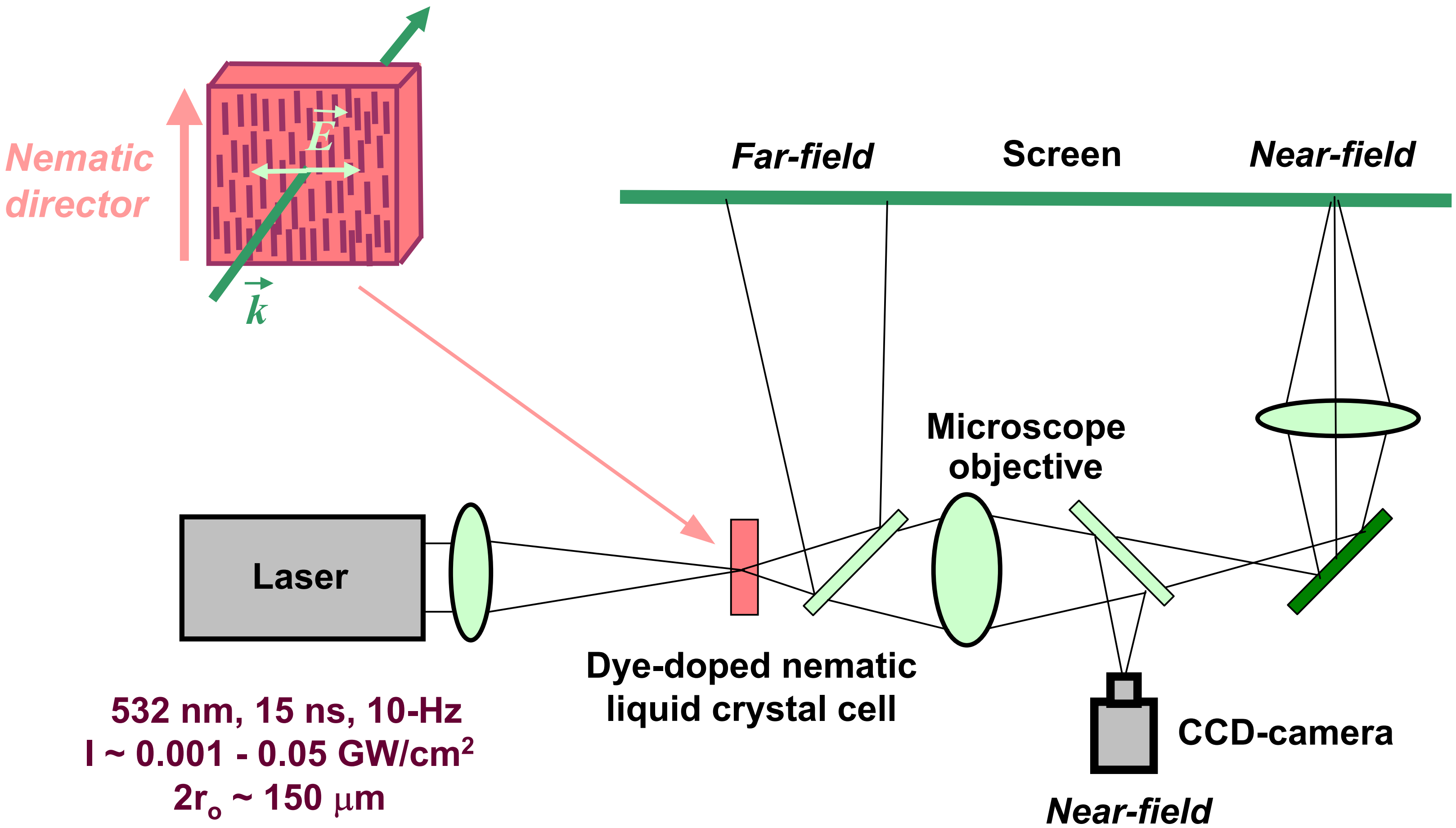
Result summary

- At I ~ 0.005 GW/cm^2, induced-birefringence effects manifest themselves in the appearance of a polarization component perpendicular to the incident polarization and parallel to the nematic director with a Maltese cross spatial pattern in the far-field.

- At higher incident intensities (I ~ 0.01 – 0.05 GW/cm^2), a second pattern-formation mode is observed and only for perpendicular to the nematic director polarization component. A high-definition, feedback-free far-field pattern changes with pulse-repetition rate from rings and stripes to multiple hexagons.

- Memory effect (frozen-in molecular reorientation) is observed. Probe, ~1 mW cw-laser beam reads multiple hexagon spatial pattern in the far-field after nanosecond laser switching off.

Experimental set up



What are liquid crystals?

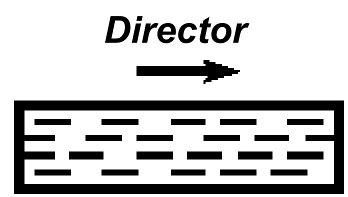
Liquid crystal is intermediate phase (mesophase) between crystalline solid and isotropic liquid

In the *nematic* phase *anisotropic* rod-like liquid crystal molecules oriented preferably in one direction (director).

While in this phase, the long axis of liquid crystal molecules can be uniformly aligned both parallel and/or perpendicular to the fluid container's walls, by special surface treatment

Nematic phase

Isotropic phase



Planar-aligned nematic layer

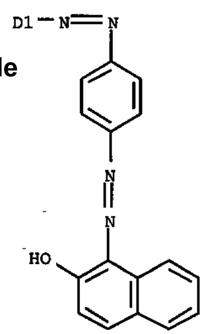
When heated nematic liquid crystals undergo a phase transition from the *nematic* to the *isotropic* (randomly oriented) phase

Liquid crystal cell preparation

Nematic liquid crystal mixture E7 doped with dye “Oil Red O” (1.5% weight-concentration) with dichroic properties was used

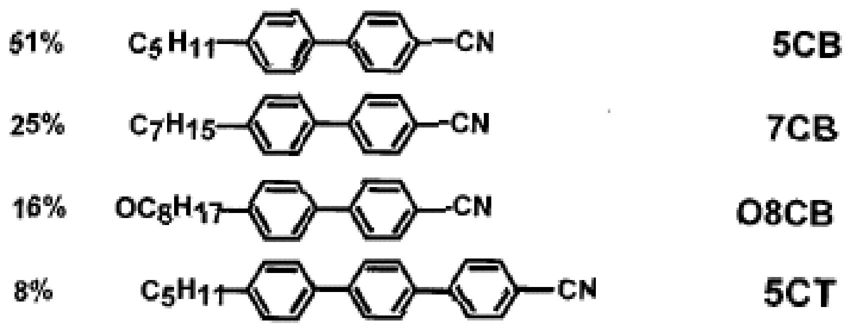
- Planar-aligned nematic liquid crystal layers were prepared using buffing techniques on Nylon 6/6 alignment layers;
- Cell thickness was ~ 10 - 20 μm;
- Cell transmittance at low incident intensities:
  1. ~ 1% for incident polarization perpendicular to nematic director;
  2. ~ 10 - 15 % for parallel polarization.

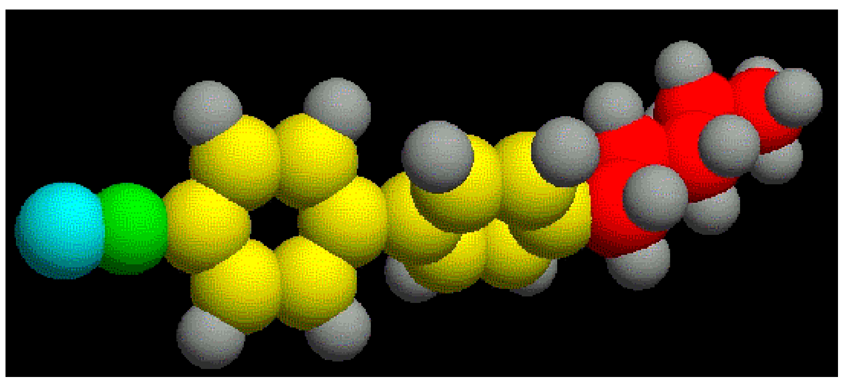
D1 = Me



Molecular structure of a dye “Oil Red O”

Liquid crystal cell preparation (continued): E7 nematic mixture composition





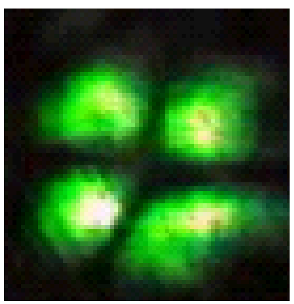
Space-filling model of 5CB (the main component of E7)

Results: Induced-birefringence effects for incident polarization perpendicular to nematic director

*S*patial pattern formation at low incident intensities (I ~ 0.005 GW/cm^2, 10-Hz prr)

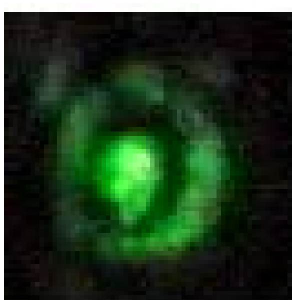
- **During repetitive illumination by a focused laser beam over timespans of *several seconds* to *several minutes*, we observed a stable far-field pattern of a "cross" whose bright axes are oriented at 45^o to the incident polarization.**

||



- **A parallel-polarization component appeared in a beam after passing through the nematic layer. Parallel polarization component evolves into the far-field in a form of a Maltese cross (optical four-leaf clover).**

⊥

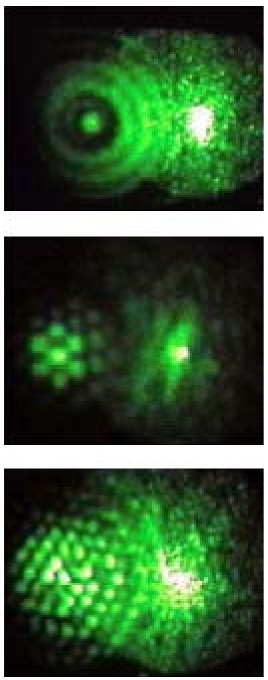


- **The incident, perpendicular polarization component evolves into the far-field pattern in a form of ring with a bright spot inside.**

Results: Spatial pattern formation at high incident intensities (I ~ 0.01 GW/cm^2, 10-Hz prr).

High-definition patterns were observed only for polarization component perpendicular to nematic director

$\perp$ $\parallel$

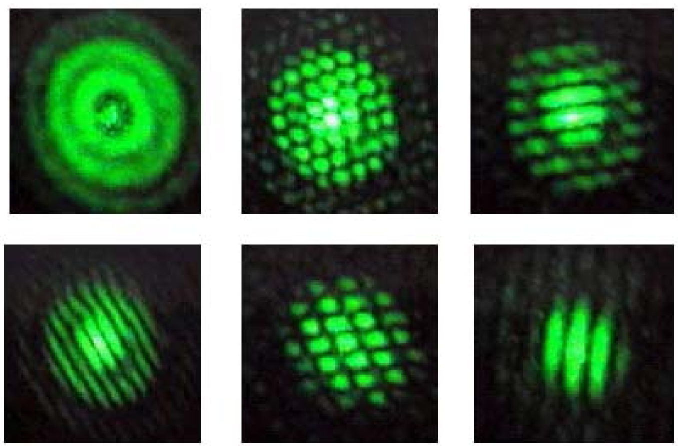


- **Figures show the beam cross-sections of two partially separated polarization components overlapping in the center: the left side of each image depicts *the perpendicular-polarization* component, the right side – *the parallel-polarization* component.**
- **The "Maltese cross" of *parallel-polarization* component (right side) smears into a random speckle pattern with a bright spot in the center.**
- **At the same time, *the perpendicular-polarization* component develops a high-definition pattern that kaleidoscopically changes from pulse to pulse from rings and stripes to multiple hexagons.**

Figures show random selection of the far-field patterns at the same incident intensity. For visualization of some details, the image in the center was recorded through an absorbing filter.

Results: Spatial pattern formation at high incident intensities (continued), I ~ 0.01 GW/cm^2, 10-Hz prr

Feedback-free far-field kaleidoscope of patterns for polarization component, perpendicular to nematic director (incident polarization)



Random selection of the far-field patterns at the same incident intensity

***Angular dimensions*:**

$\theta_o = 8.10^{-3} - 2.10^{-2}$ for highest spatial frequencies of hexagons and stripes;

$\theta_\alpha = 4\,.10^{-2} - 1.3.10^{-1}$ for divergence cone of the whole beam.

Calculated size of near-field inhomogeneities $d = 1.22\lambda/\theta$:

$d_o = 32 - 81\ \mu m$; $d_\alpha = 5 - 16\ \mu m$.

Results: Spatial pattern formation at high intensities (continued), I ~ 0.01 GW/cm^2, 10-Hz prr

The key characteristics of kaleidoscope-pattern-formation phenomenon

1. The pattern phenomenon has a threshold I_{thr} that depends on the cell transmittance (I_{thr}~ 0.005 GW/cm^2)

2. The effect is cumulative in nature at I ~ 0.005 ~ 0.05 GW/cm^2. Pattern mode has a buildup time of several seconds to minutes depending on the incident intensity and cell transmittance. Strong scattering and a wide-spot pattern in the far-field afterwards manifest the beginning of a kaleidoscope-pattern-mode.

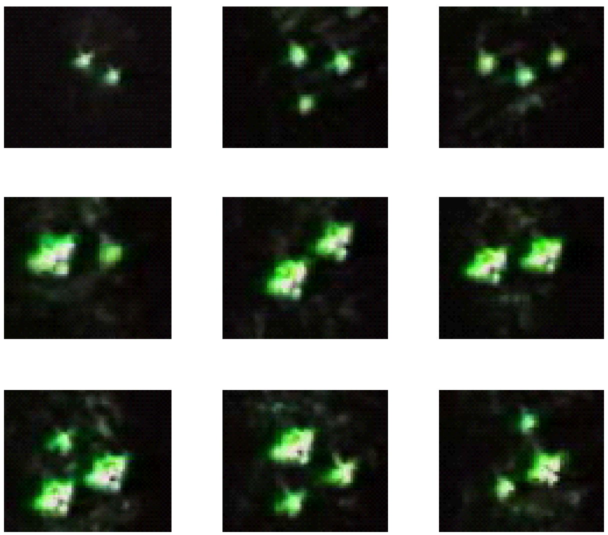
3. Above threshold, the kaleidoscope of patterns exists for all time of irradiation (we observed these patterns for hours), stops with switching off the laser, and recoveres again upon switching it on.

4. Rotating the planar-aligned cell around the light-propagation direction changes the threshold.

5. The effect disappears after switching the laser from a 10-Hz to a 5-Hz repetition-rate mode.

Results: Spatial pattern formation at high incident intensities (continued), I ~ 0.01 GW/cm^2, 10-Hz prr

Near-field images of intensity distribution at 500 x magnification recorded from the screen



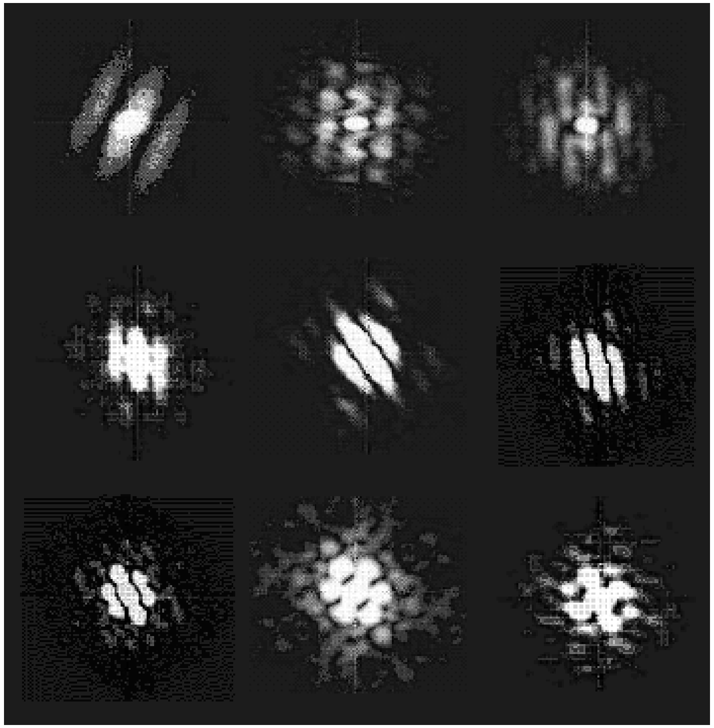
Random selection of the near-field patterns at the same incident intensity

CCD-images showed the size of the spots d_α ~ 5 - 15 μm with distance between spots d_o ~ 35 - 70 μm.

These values of d_α and d_o agree well with evaluations from the observed far-field angular dimensions θ_o and θ_α

Results: Spatial pattern formation at high incident intensities (continued), I ~ 0.01 GW/cm^2, 10Hz prr

Numerical modeling of the far-field intensity distribution from the near-field images



Electric field distribution in the far-field

$$\mathbf{E}_2(x,y) = \frac{K}{z} e^{ikz} e^{(ik/2z)(x^2+y^2)} \mathbf{F}[\mathbf{E}_1(\zeta,\eta)\mathbf{A}(\zeta,\eta)]$$

for large z, where $\mathbf{F}$ denotes Fourier transform ; $\mathbf{E}_1$ is the electric field at the aperture and $\mathbf{A}(\zeta,\eta)$ is an aperture function with aperture radius $\mathbf{R}$:

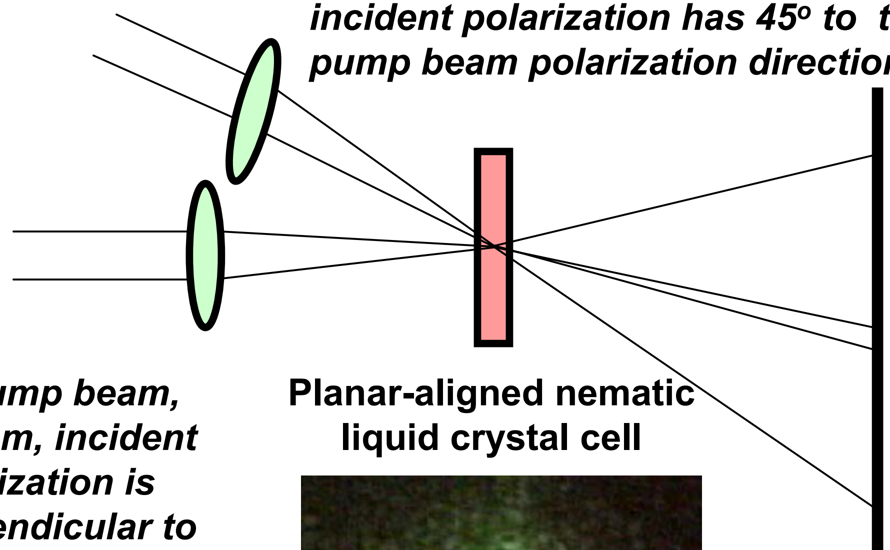
$$\mathbf{A}(\zeta,\eta) = \begin{cases} 1, & \zeta^2 + \eta^2 \le R^2 \\ 0 & \text{otherwise} \end{cases}.$$

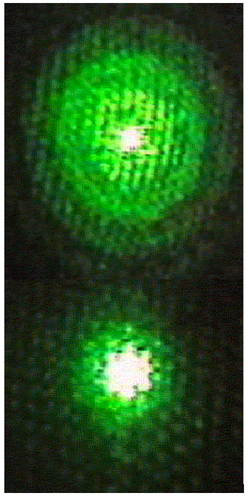
Calculations were made in approximation of constant phase along all spots

Results: Spatial pattern formation at high incident intensities (continued), I ~ 0.01 GW/cm^2. Memory effect

Probe, ~ 1 mW cw laser beam reads the multiple-hexagon spatial pattern in the far-field for hours after a ns-pump beam switching off.

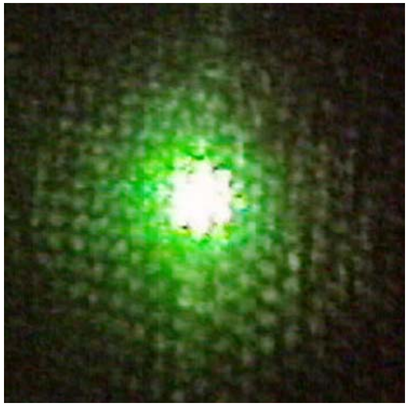
Cw weak probe beam, 532 nm, incident polarization has 45^o to the pump beam polarization direction





ns-pump beam, 532 nm, incident polarization is perpendicular to nematic director

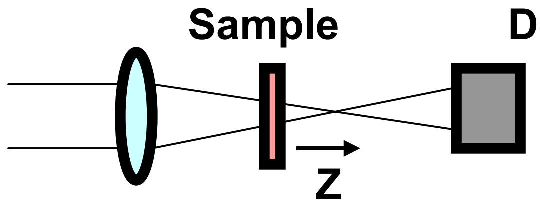
Planar-aligned nematic liquid crystal cell

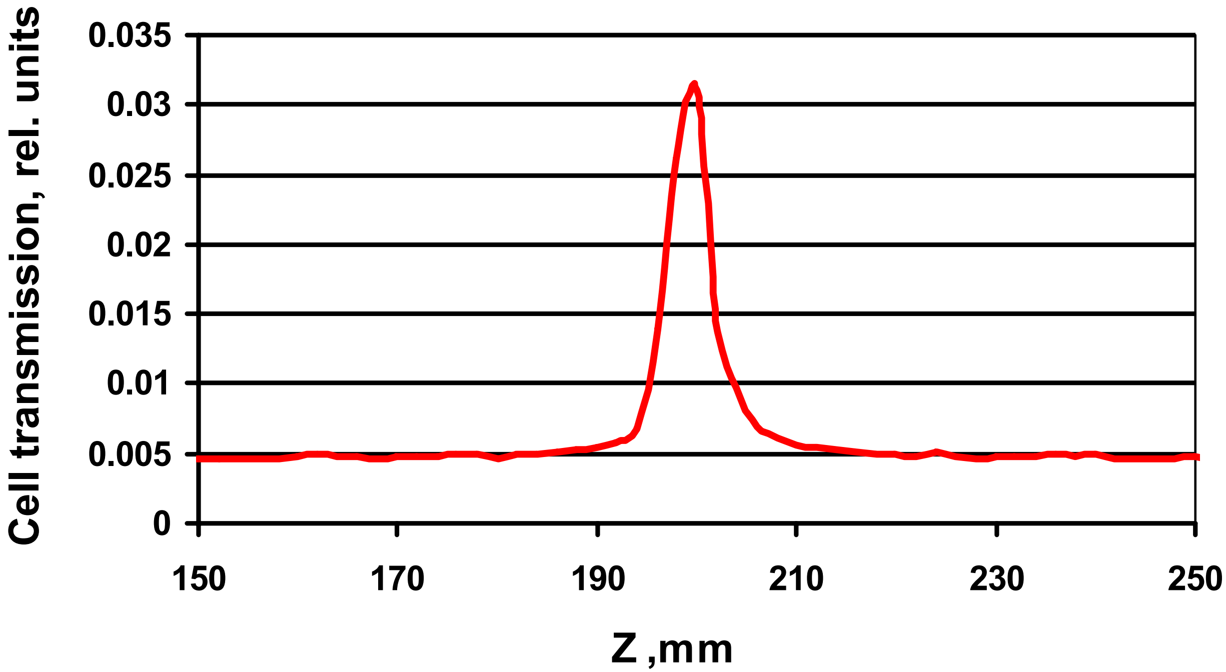


Far-field pattern of the probe beam 30 min after the pump beam was switched off.

Results: Nonlinear transmission enhancement of a dye-doped liquid crystal layer at below threshold incident intensities

Z-scan measurements showed several times enhancement of the cell transmittance.





Discussion: Laser-induced birefringence, low-intensity-pattern-formation in a form of a Maltese cross and memory effect (I ~ 0.005 GW/cm^2, 10-Hz prr)

The mechanism of a heat-flow birefringence caused by a locally inhomogenious temperature field is a likely explanation for the induced-birefringence experiment and low-intensity-pattern formation (appearance of Maltese cross in beam cross-section).

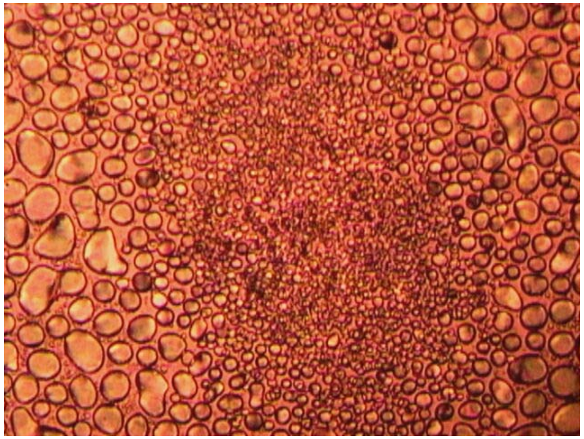
Light absorption and laser-induced, local heating cause molecular reorientations of liquid crystal and rotations of the optical axis. Observed birefringence patterns and reorientation phenomena can be explained by stress-optical coupling where the stress is caused by a locally inhomogeneous temperature field [3].

3. R. Elscher, R. Macdonald, H.J. Eichler, S. Hess, A.M. Sonnet, "Molecular reorientation of a nematic glass by laser-induced heat flow", *Phys. Rev. E*, 60, 1792- 1798 (1999).

Discussion: How do hexagonal patterns emerge from a Gaussian initial spatial intensity distribution?

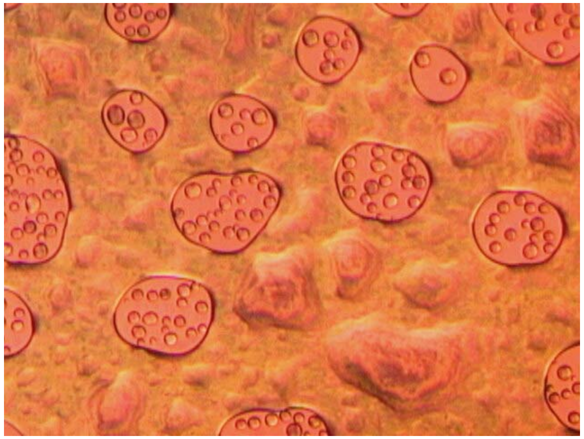
Optical microscope images of a phase nucleation in a dye-doped nematic mixture E7 near the nematic/isotropic-liquid phase transition (T = 58^o C)

In this experiment nematic E7 was heated inside a Mettler hot stage with 0.1^{o}C heating steps. No laser radiation was used.



1mm

Heating



1 mm

Cooling

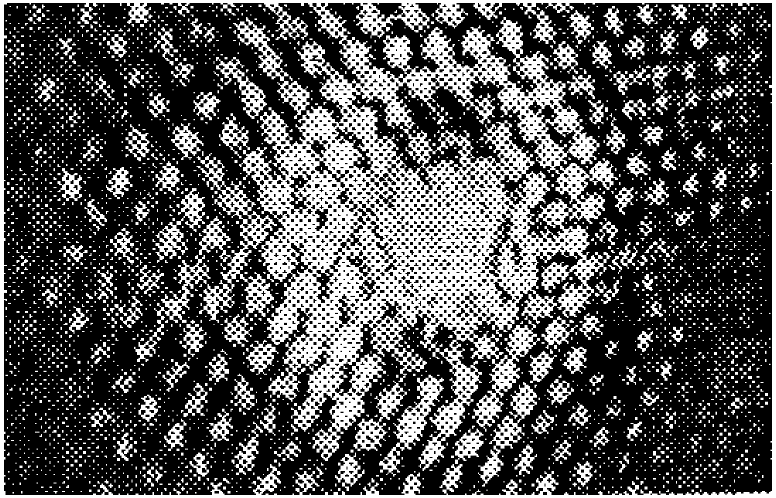
Drops of isotropic liquid with sizes between several and hundred microns exist inside the nematic material at ΔT = 1 - 2^{o}C below nematic/isotropic phase transition

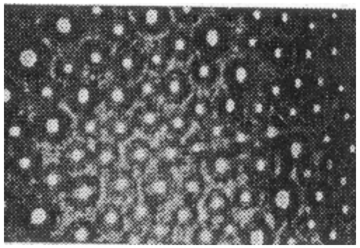
Discussion: How do hexagonal patterns emerge from a Gaussian initial spatial intensity distribution (continued)?

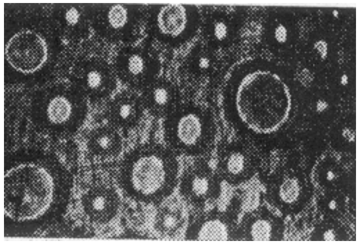
- **Laser driven nematic liquid crystal/isotropic-liquid phase transition with appearance of phase-transition-drops of isotropic liquid in nematic *mixture* and nonlinear enhancement of transmission of a dichroic dye are apparently responsible for this effect.**
- **The patterns' ring structure can be attributed to the diffraction of laser light at the sharp edge of drops of isotropic liquid. The variety of drop numbers in focus, their size and the distance between them, and a gradient of transmittance inside the drop define enormous variety of patterns we observed.**
- **The absence of hexagonal patterns for *parallel polarization* can be explained by the much lower difference in absorption of a dichroic dye in a nematic/isotropic state for parallel polarization in comparison with perpendicular polarization.**

Discussion: How do hexagonal patterns emerge from a Gaussian initial spatial intensity distribution (continued)?

Existence of phase nucleation near nematic/isotropic phase transition of undoped nematic *mixtures* in a form of multiple isotropic drops were reported by A.S. Zolot'ko and V.F. Kitaeva (JETP Lett., 62, 124 (1995))







Diffraction of a low power density, cw probe beam on isotropic drops created *by heating in the oven* to the phase-transition-temperature showed a far-field small-scale hexagonal patterns similar to some of hexagonal patterns observed in our experiments

Summary

- **The interaction of ~ 15-ns, 10-Hz prr, 532-nm laser beam with a planar-aligned, dye-doped nematic liquid crystal layer has been studied for incident polarization perpendicular to nematic director.**

- ***Cumulative* induced-birefringence effects were observed. They manifest themselves in (1) appearance of a polarization component parallel to nematic director and (2) two modes of spatial pattern formation with different patterns for parallel and perpendicular polarizations.**

- **At low-intensity mode (I ~ 0.005 GW/cm^2) spatial birefringence patterns are different for each polarization, e.g., a Maltese cross was observed in laser-induced (parallel to nematic director) polarization component. At the same time a central bright spot with a ring appeared in an incident (perpendicular) polarization component.**

- **The observed birefringence with Maltese cross spatial pattern can be explained by laser-induced heat flow and stress, leading to molecular reorientation which is then frozen if the optical excitation is switched off. Accumulation of molecular reorientation from pulse to pulse gives a stable pattern formation in a laser beam.**

Summary (continued)

- **High-intensity-mode (I ~ 0.01 - 0.05 GW/cm^2) spatial pattern formation has also a polarization asymmetry. E.g., high-definition patterns were observed only for polarization component perpendicular to nematic director. Highly reproducible and easy to handle pattern formation manifests itself in kaleidoscopic change of pattern with laser pulse repetition rate from rings and stripes to multiple hexagons.**
- **Laser driven nematic liquid crystal/isotropic liquid phase transition with appearance of phase-transition-drops of isotropic liquid in nematic *mixture* and nonlinear enhancement of transmission of a dichroic dye are apparently responsible for high-intensity hexagon/stripes spatial patterns in initial Gaussian laser beam.**
- **Memory effect (frozen-in molecular reorientation) is observed. Probe, cw, 1mW laser beam reads multiple hexagon spatial pattern in the far-field after nanosecond laser switching off.**

Acknowledgements

The work was supported by the U.S. Department of Energy Office of Inertial Confinement Fusion under Cooperative Agreement No. DE-FC03-92SF19460, the University of Rochester, and the New York State Energy Research and Development Authority. The support of DOE does not constitute an endorsement by DOE of the views expressed in this article. The authors also acknowledge the support by the Office of Naval Research and the Army Research Office. The authors thank B. Watson, B. Klehn, and D. Hurley for preparation of some of the liquid-crystal cells, Dr. N. Lepeshkin for the assistance in Z-scan measurements, G. Piredda for the help in numerical modeling, and Dr. A. Schmid and R. Bennink for numerous fruitful discussions.