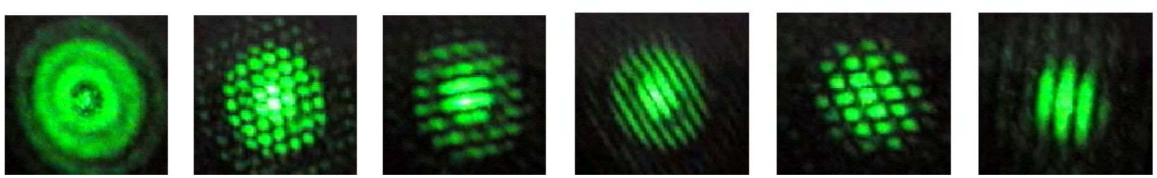
Feedback-free kaleidoscope of patterns from nanosecond laser irradiated nematic liquid crystals

Svetlana G. Lukishova, Robert W. Boyd, Nick Lepeshkin and Ryan S. Bennink
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

Generally optical feedback is necessary for hexagonal pattern formation in nonlinear optics

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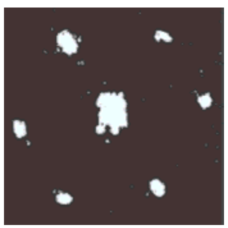
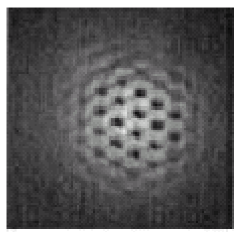
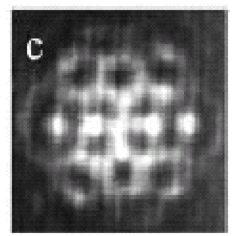
1. M.A. Vorontsov and W.B. Miller, Eds., *Self-Organization in Optical Systems and Applications in Information Technology,* Springer (1985).
2. Transverse Effects in Nonlinear Optical Systems, Special issues of *J. Opt. Soc. Am*. B7, is. 6 and 7 (1990) with overview of N.B. Abraham and W.J. Firth.



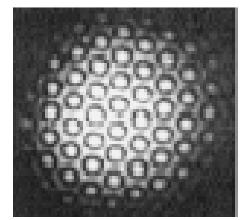
Grynberg et al. (1986)



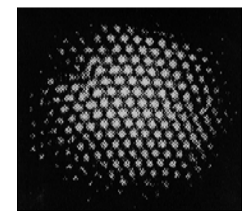
Ackerman et al.(1995)



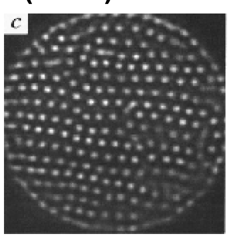
Vaupel et al. (1999)



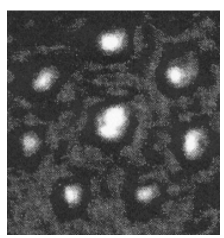
Luchnikov et al. (1999)



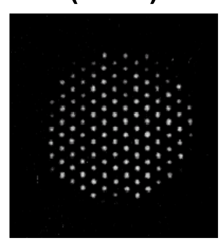
Banerjee et al. (1995)



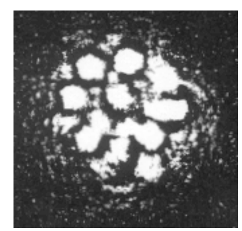
Vorontsov et al. (2000)



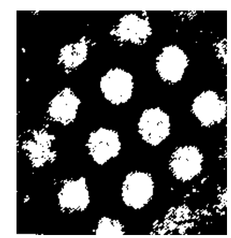
Arecchi et al. (1994)



Neubecker et al. (1995)

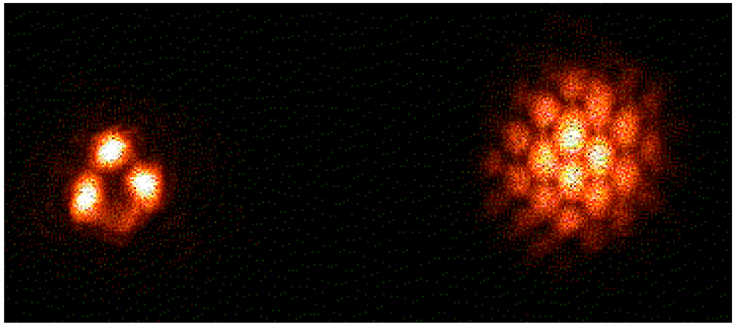


Macdonald et al. (1992)



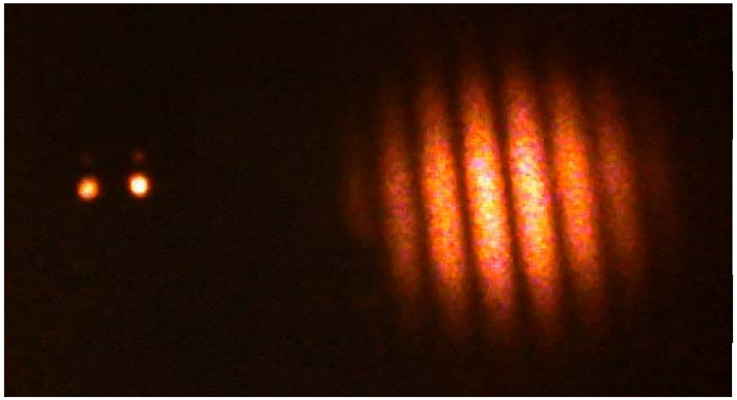
Tamburrini et al. (1993)

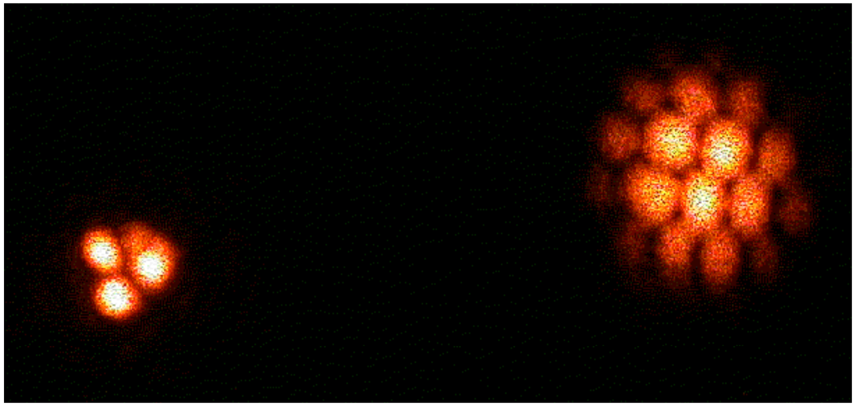
The purpose of this paper is to show hexagonal pattern formation in a feedback-free nonlinear optical system



Feedback-free hexagonal (honeycomb) pattern formation was reported recently in atomic sodium vapor

Bennink R. et al., *PRL*, 88 (11) 113901 (2002)





Feedback-free kaleidoscope of patterns from nanosecond laser irradiated highly-absorbing nematic liquid crystal

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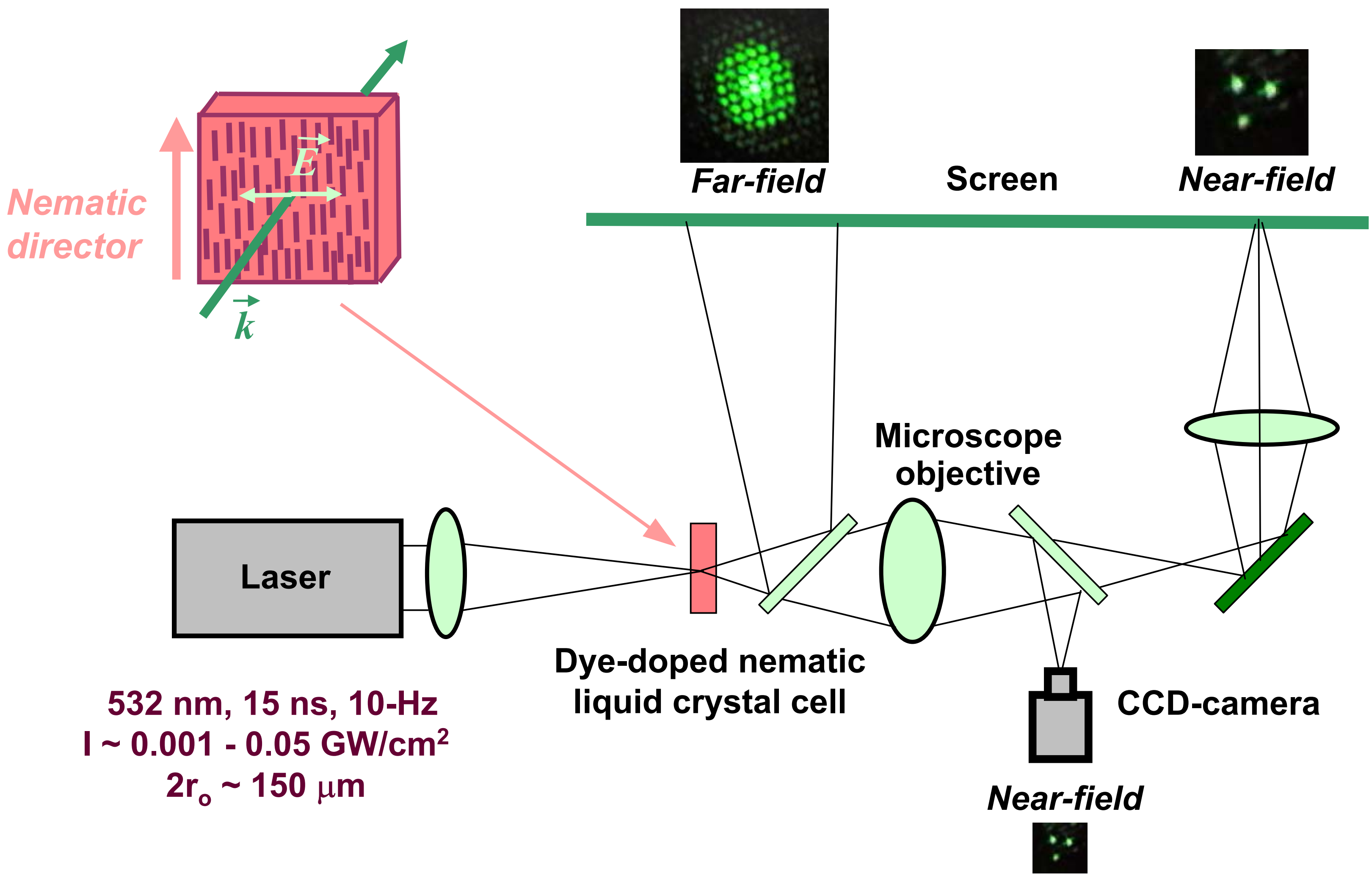
Content:

- Experimental set up
- Dye-doped nematic liquid crystal cells
- Far-field feedback-free hexagonal pattern formation
- Near-field patterns
- Memory effect
- Z-scan measurements of nonlinear transmission of dye-doped nematic liquid crystal layers
- Mechanism of the phenomenon
- Summary

Experimental set up



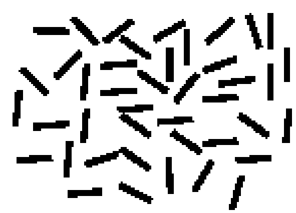
What are liquid crystals?

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

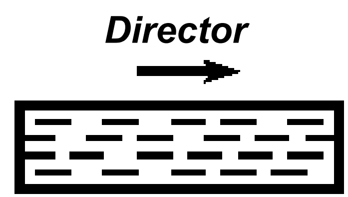
In the *nematic* phase *anisotropic* rod-like liquid crystal molecules oriented preferably in one direction (director).

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



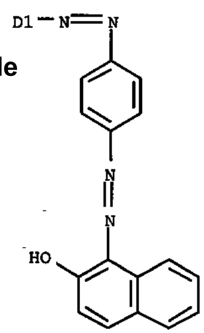
Planar-aligned nematic layer

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

Liquid crystal cell preparation

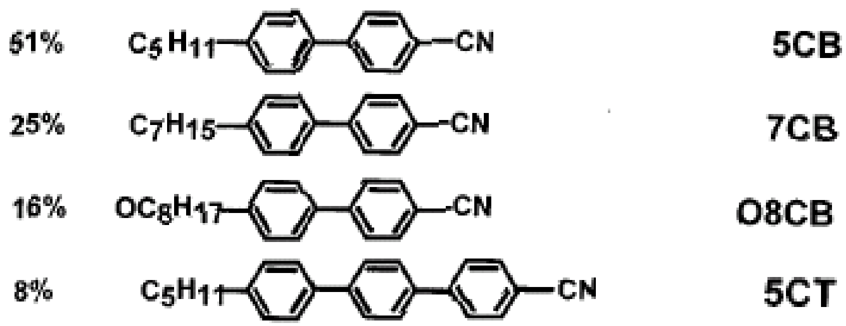
Nematic liquid crystal mixture E7 doped with dye “Oil Red O” (1.5% weight-concentration) with dichroic properties was used

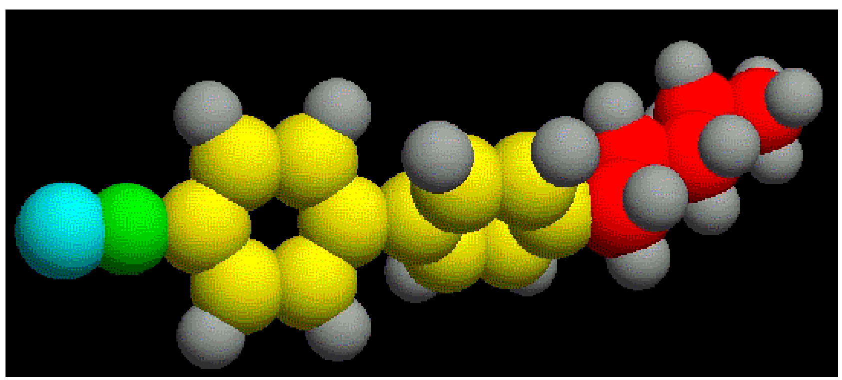
- **Planar-aligned nematic liquid crystal layers were prepared using buffing techniques on Nylon 6/6 alignment layers;**
- **Cell thickness was ~ 10 - 20 μm;**
- **Cell transmittance at low incident intensities:**
  1. **~ 1% for incident polarization perpendicular to nematic director;**
  2. **~ 10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



Molecular structure of a dye “Oil Red O”

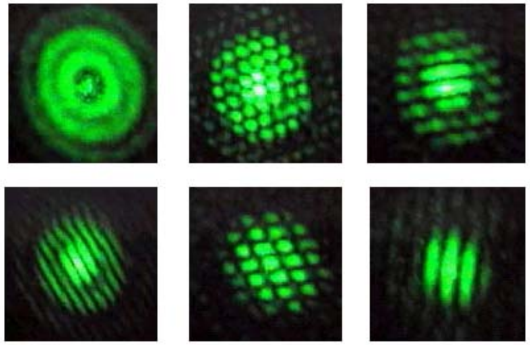
Liquid crystal cell preparation (continued): E7 nematic mixture composition



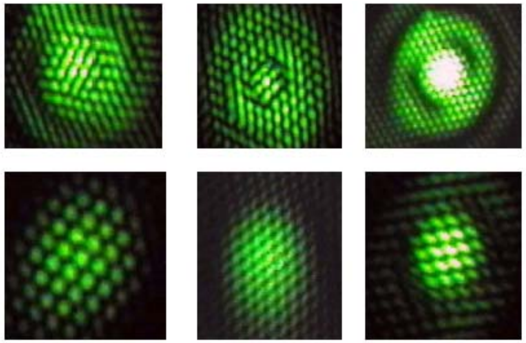


Space-filling model of 5CB (the main component of E7)

Feedback-free kaleidoscope of patterns: far-field



50-cm from the output of planar-aligned cell



50-cm from the output of unaligned cell

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

***Angular dimensions*:**

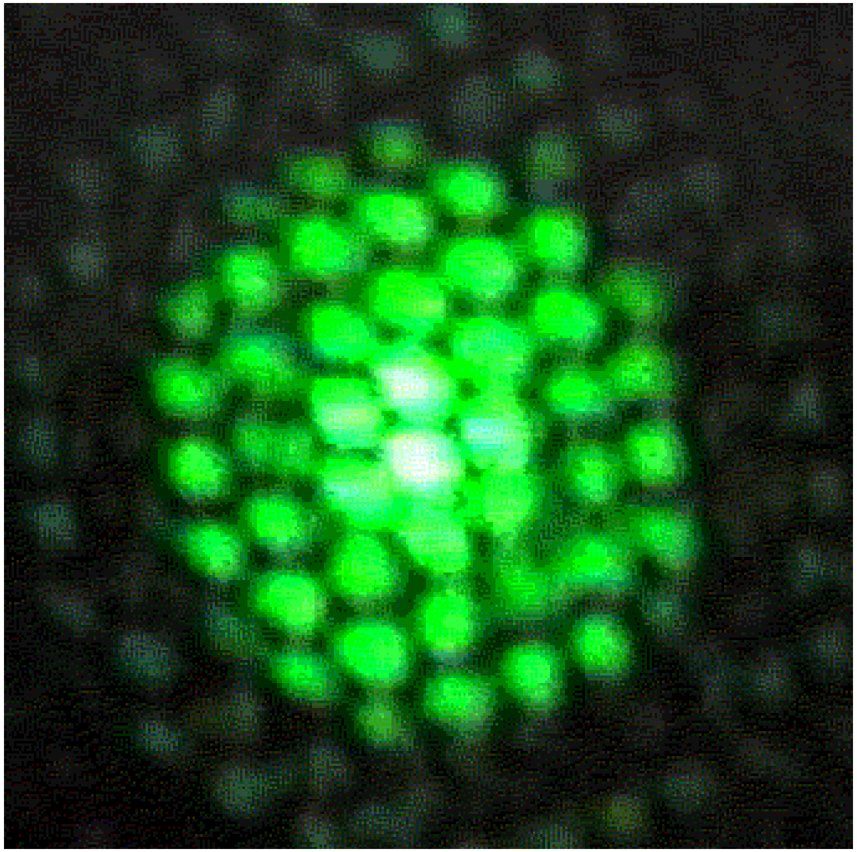
θ_o = 8.10^{-3} - 2.10^{-2} for highest spatial frequencies of hexagons and stripes;

θ_α = 4 .10^{-2} - 1.3.10^{-1} for divergence cone of the whole beam.

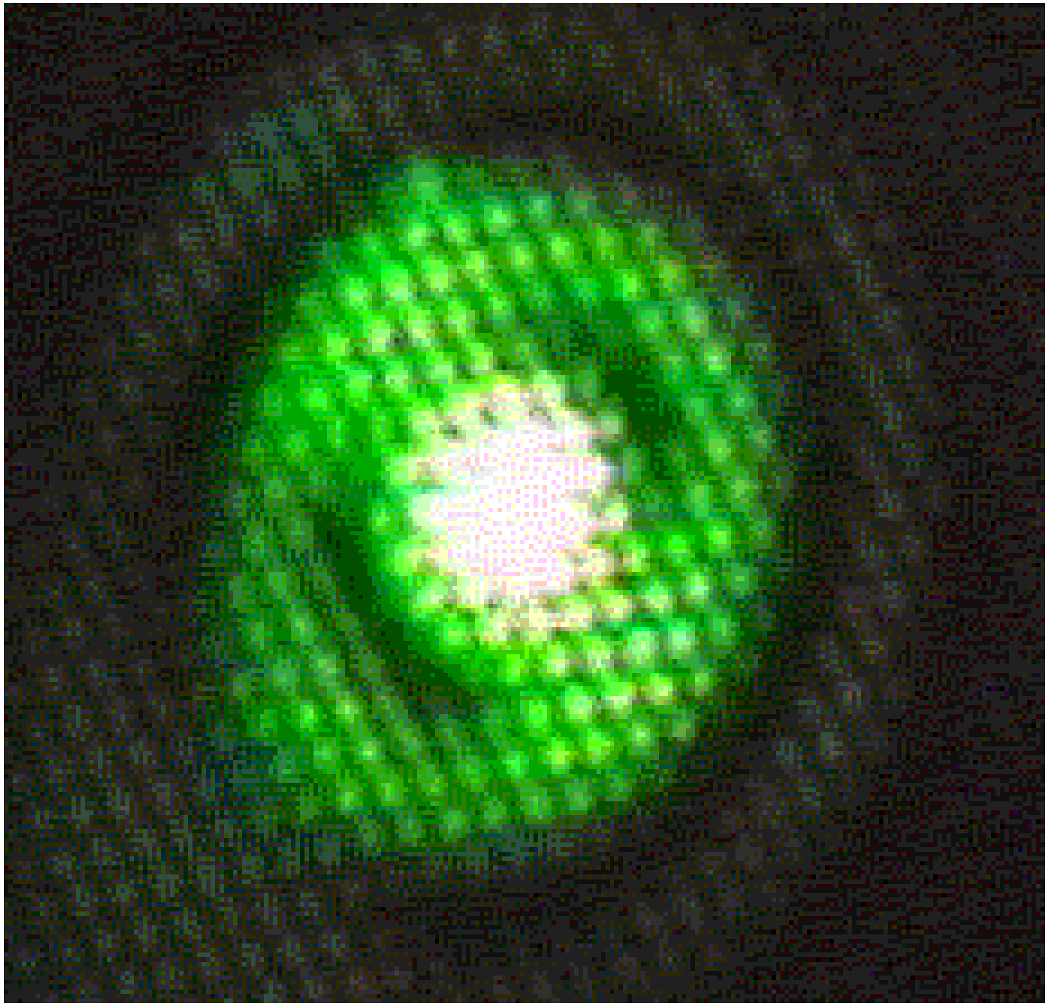
***Calculated size of near-field inhomogeneities* d = 1.22λ/ θ:**

d_o = 32 - 81 μm; d_α = 5 - 16 μm.

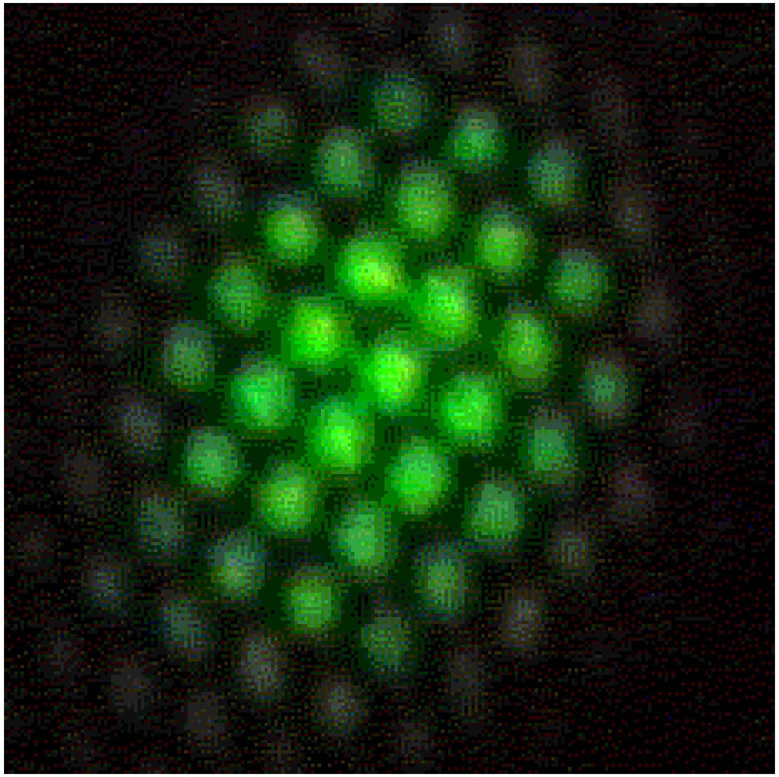
Hexagons in the far-field



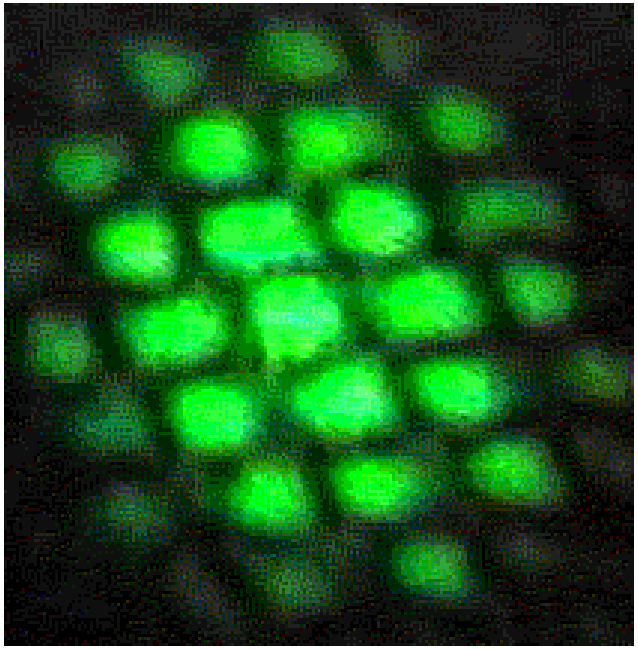
Hexagons in the far-field (continued)



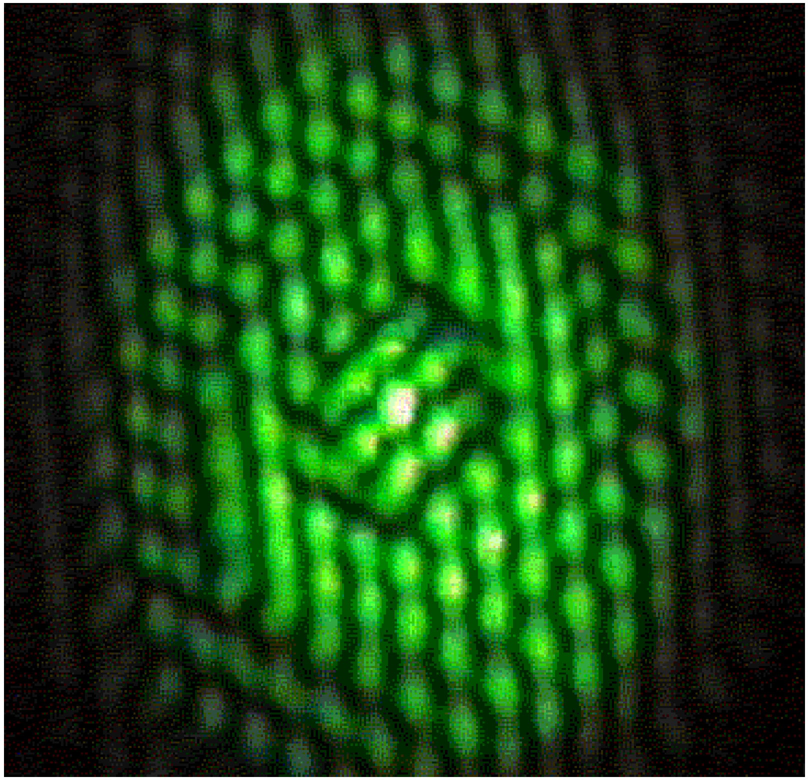
Hexagons in the far-field (continued)



Hexagons in the far-field (continued)



Hexagons in the far-field (continued)



The key characteristics of kaleidoscope-pattern-formation phenomenon (I ~ 0.01 GW/cm^2)

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1. Patterns were recorded for both planar-aligned and unaligned cells.
2. The pattern phenomenon has a threshold that depends on the cell transmittance (I_{thr}~ 0.005GW/cm^2).
3. The effect is <u>cumulative</u>. Pattern mode has a buildup time of several seconds to minutes depending on the incident intensity.
4. Strong scattering with a sharp increasing of a beam diameter and appearance of rings in the far-field manifest the beginning of a kaleidoscope-pattern-mode.
5. Above threshold, we observed kaleidoscope of patterns for hours.
6. Rotating the planar-aligned cell around the light-propagation direction changes the threshold.
7. The patterns disappeared after switching the laser from a 10-Hz to a 5-Hz repetition-rate mode at I ~ 0.01-0.05 GW/cm^2.

Feedback-free kaleidoscope of patterns: near-field

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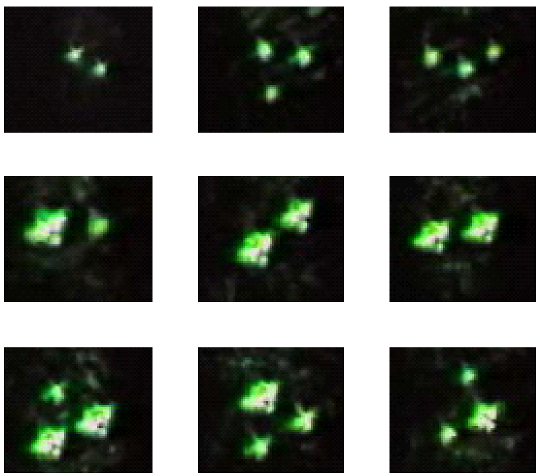
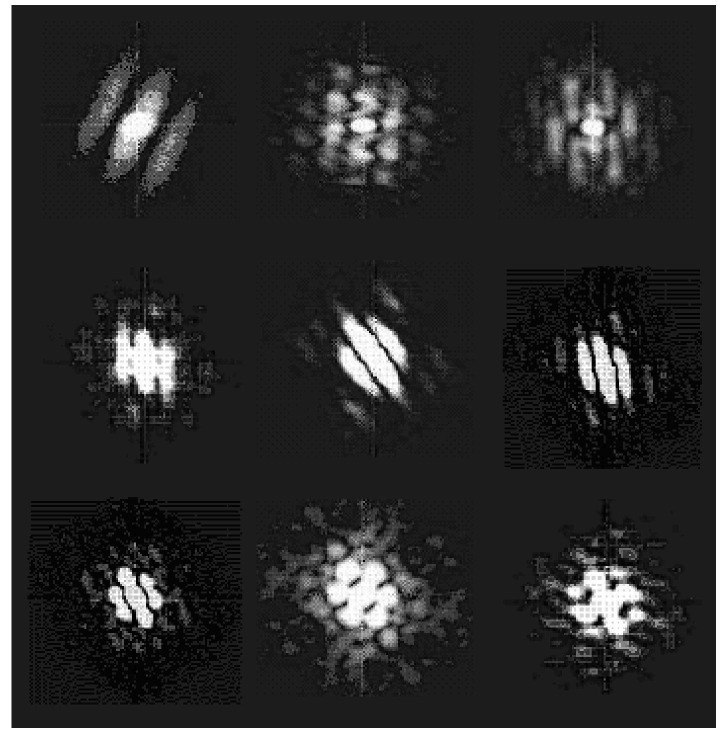
Random selection of the near-field patterns at the same incident intensity



500 x magnification



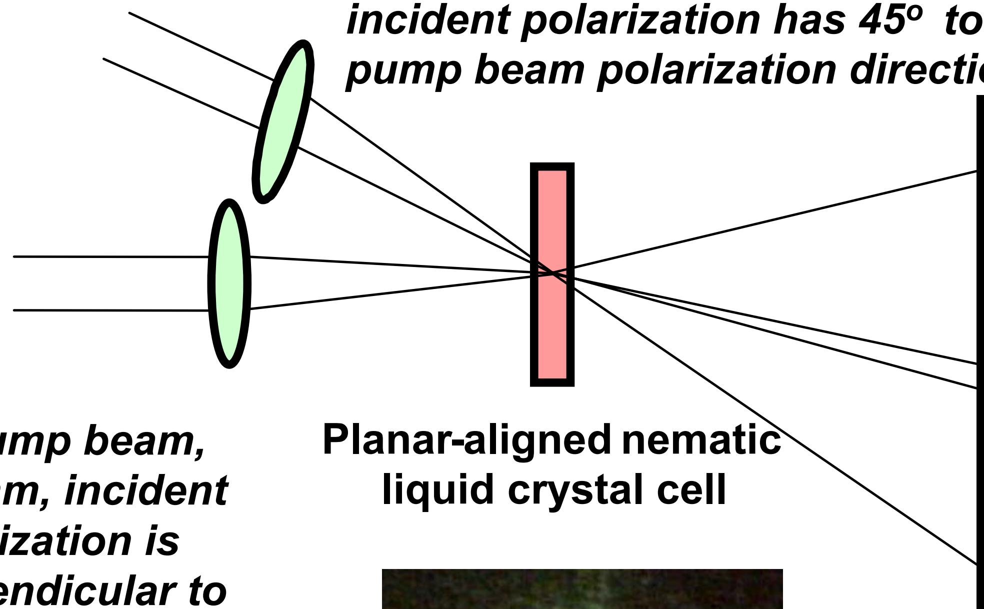
The size of the spots $d_\alpha \sim 5 - 15\ \mu m$ with distance between spots $d_o \sim 35 - 70\ \mu m$.

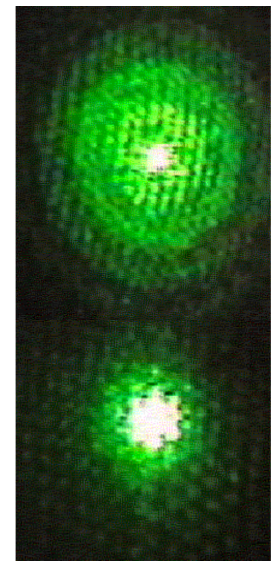
Calculated from the far-field *experiments* $d_\alpha = 5 - 16\ \mu m$; $d_o = 32 - 81\ \mu m$.

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

<u>Memory effect</u>: Probe, ~ 1 mW cw laser beam reads the multiple-hexagon spatial pattern in the far-field for hours after a nanosecond-pump beam switching off.

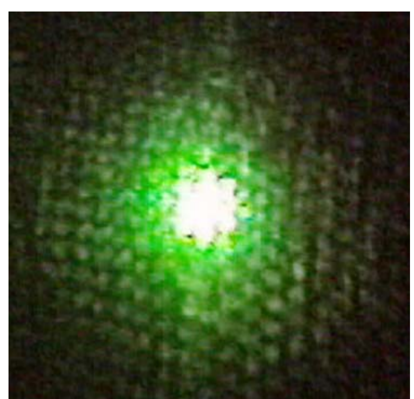
Cw weak probe beam, 532 nm, incident polarization has 45º to the pump beam polarization direction





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.

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

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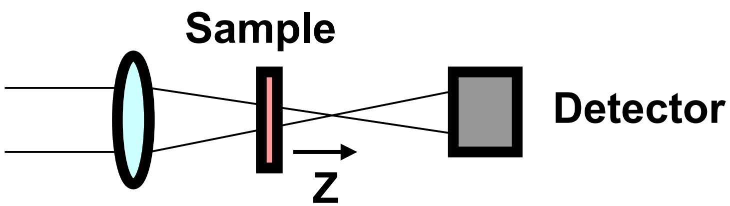
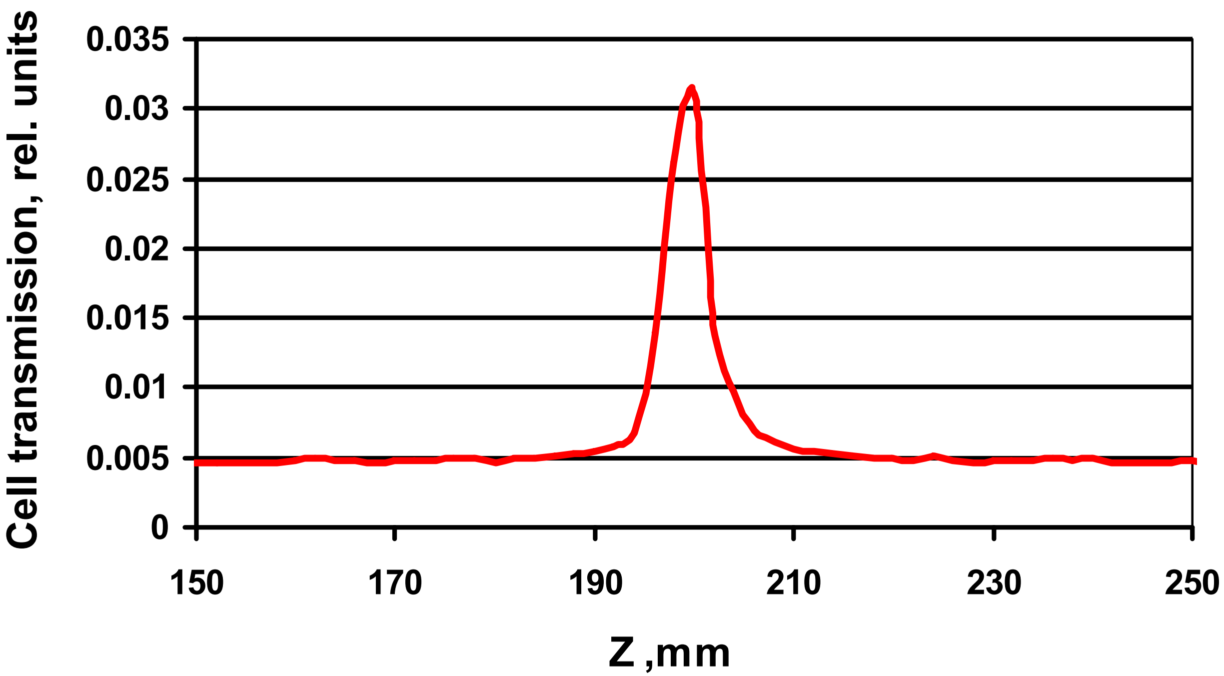
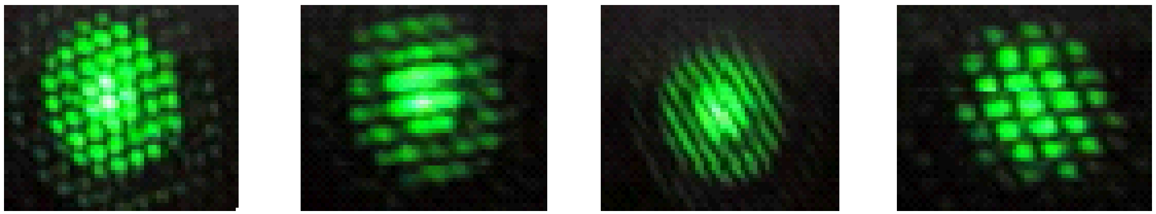
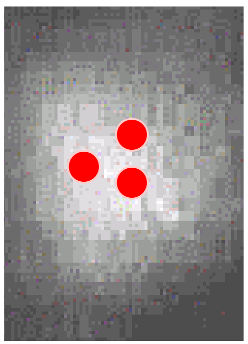
Z-scan measurements showed several times enhancement of the cell transmittance.





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





Laser driven nematic/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.

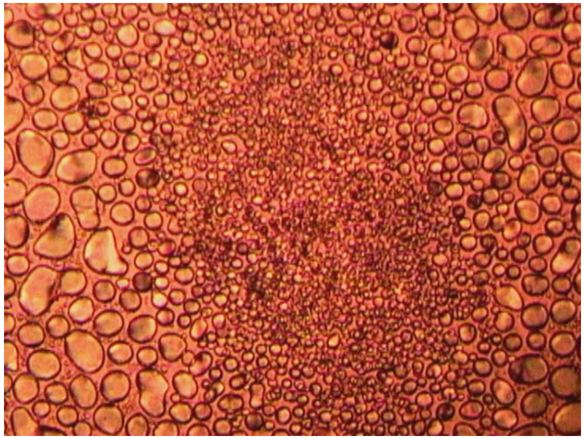
Laser-beam cross-section

•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.

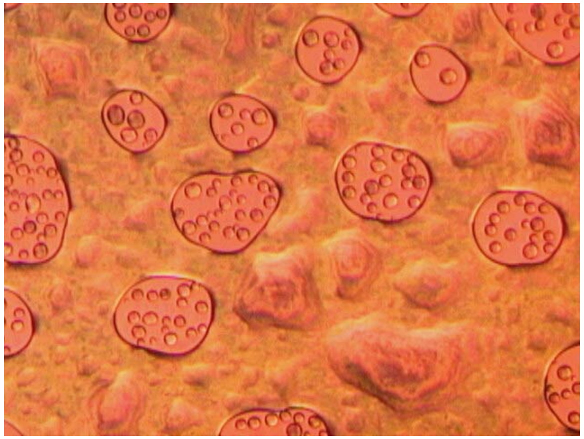
Optical microscope images of a phase nucleation in a dye-doped nematic mixture E7 near the nematic/isotropic-liquid phase transition (T = 58^{o} C)

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



1mm

Heating

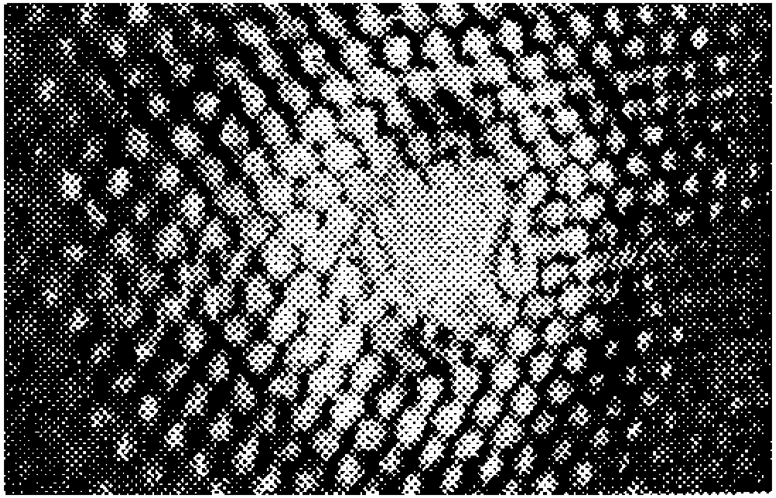


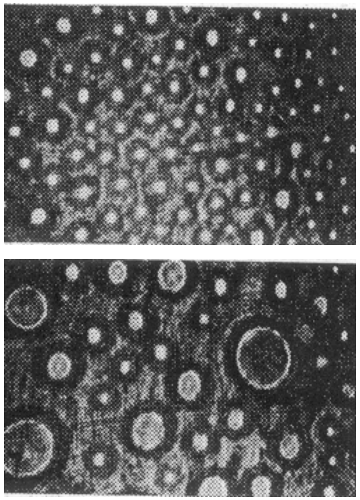
1 mm

Cooling

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

Existence of phase nucleation near nematic/isotropic phase transition of <u>undoped</u> 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

Why do cumulative effects exist in laser heating of thin, 10 – 20 μm layers, at 10-Hz prr?

1. Maximum instantaneous temperature during the single pulse $T_{inst} = T_{room} + E/\rho Shc_p \sim 100^oC$, where absorbed energy E = 25 μJ; density $\rho = 1 g/cm^3$; cross-section $S = \pi(75\ \mu m)^2$; layer thickness h = 10 μm; heat capacity $c_p = 1.92$ J/gK.
2. Numerical modeling of a heat-transfer in 10 - 20 μm thickness layers between glass substrates showed heat dissipation in time interval ~ 0.5ms, much shorter than time interval between two-pulses.
3. Similar numerical modeling has been made in Ref. 1 for thin, 2 μm thickness layers, where ~10 μs dissipation time has been reported.
4. Heat dissipation in much longer intervals ~ 0.1 s was reported in Ref. 2 for *thick* ~ 100 μm layers.
5. Additional mechanism of a ***heat-isolation*** should be taken into account in our experiments to explain a *cumulative* character of observed phenomenon. Micrometer-size air-bubbles reported in Ref. 3 on the nematic/glass interface might create heat isolation.

1) D. Grebe and R. Macdonald, *J. Phys.D.,*27, 567 (1994).

2) H. Hsiung, L.P. Shi, and Y.R. Shen, *Phys. Rev*. A, 30,1453 (1984).

3) G. Eyring and M.D. Fayer, *Chem. Phys. Lett*., 98, 428 (1983).

Summary

- New phenomenon of hexagonal pattern formation from initially Gaussian spatial intensity distribution of laser beam was observed.
- Highly reproducible and easy to handle pattern formation in *a single* laser beam and *without any feedback* involved manifests itself in kaleidoscopic change of pattern from stripes to multiple hexagons of various scales.
- (1) Laser driven nematic liquid crystal/isotropic liquid phase transition with appearance of phase-transition-drops of isotropic liquid in nematic *mixture* and (2) nonlinear enhancement of transmission of a dichroic dye are apparently responsible for this effect.
- Memory effect was observed. Probe, cw, 1mW laser beam reads multiple hexagon spatial pattern in the far-field after nanosecond laser switching off.

Acknowledgements

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The authors thank B. Watson, B. Klehn, and D. Hurley for preparation of some of the liquid-crystal cells, G. Piredda for the help in numerical modeling, and Dr. A. Schmid for numerous fruitful discussions.