Liquid Crystals under High-Power Laser Irradiation

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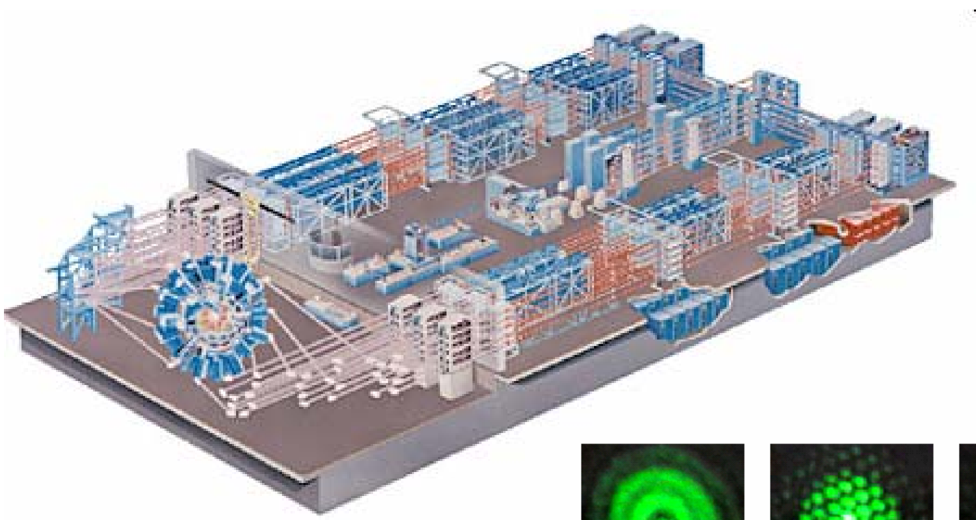
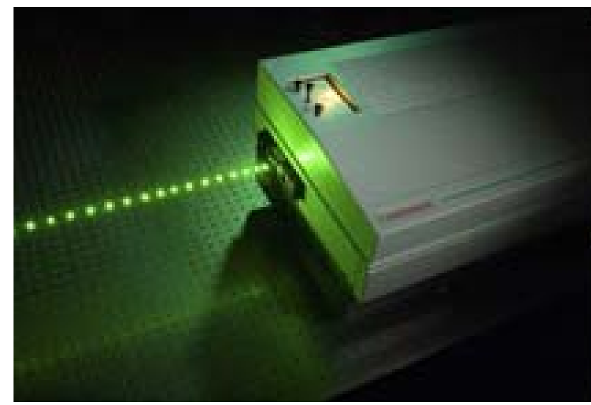
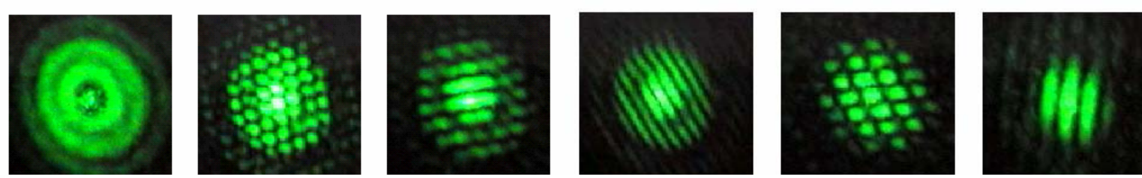
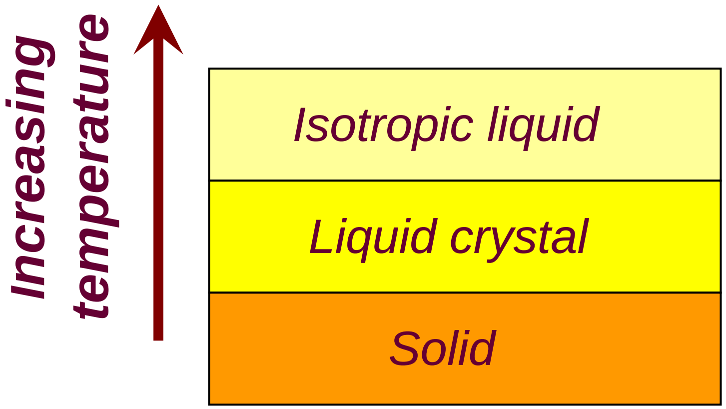


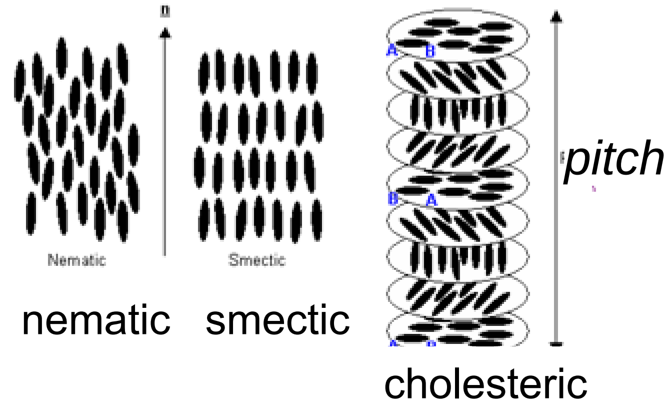




International Conference "Nonlinear Optics: East-West Reunion"
(September 21-23, Suzdal, Russia)

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





- In the nematic phase anisotropic rod-like liquid crystal molecules are oriented preferably in one direction (director).
- Chiral nematic (cholesteric) molecules self-assemble in a spiral structure.
- 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.
- When heated, thermotropic liquid crystals undergo a phase transition to the isotropic (randomly oriented) state.

Outline

- **Nonlinear optics of liquid crystals (brief overview)**
- **Our experiments on nonlinear optics of liquid crystals under nanosecond laser irradiation:**
  - nonlinear selective reflection of cholesteric photonic bandgap structures (Winful's bistability);
  - nonlinear absorption and refraction of liquid crystals;
  - feedback-free pattern formation
- **Liquid crystals lasers and single-photon sources**

Nonlinear Optics of Liquid Crystals (brief overview of some experiments)

Nonlinear Optical Effects in Liquid Crystals

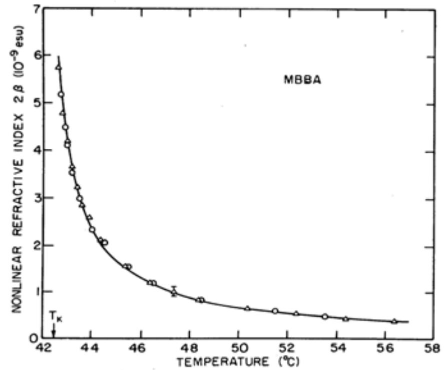
- Harmonic generation and stimulated scatterings
- Degenerate wave-mixing and phase conjugation
- Self-focusing, spatial self-phase modulation, and solitons (nematicons)
- Two-photon and excited state absorption, photoacoustic and thermal lens effects
- Optical bistability
- Optical pattern formation

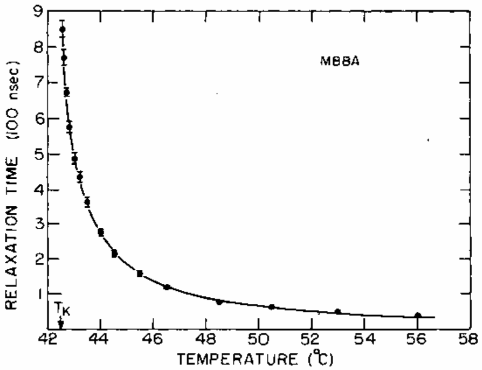
Reviews:

1. S.M. Arakelyan, G.A. Lyakhov, Yu.S. Chilingaryan, Sov.Phys.:Uspekhi 131, 3(1980).
2. S.M. Arakelyan, Yu.S. Chilingaryan, Nonlinear Optics of Liquid Crystals (1987).
3. I.C. Khoo and Y.R. Shen, Opt. Eng. 24, 579 (1985).
4. N.V. Tabiryan, A.V. Sukhov, B. Ya. Zeldovich, Mol.Cryst.Liq.Cryst. 136, 1 (1986).
5. I.-C. Khoo, Liquid Crystals, Wiley (2007)
6. S.G. Lukishova, J. Nonl. Opt. Phys. Mat. 9, 365 (2000).

The self-focusing experiments in liquid crystals date back to the 1970s. It is an ideal medium for the study of transient self-focusing.

- Dependence of n_2 and relaxation time of liquid crystal MBBA in isotropic state on the temperature (T_k is a nematic/isotropic phase transition temperature [1].





[1]. Wong and Shen, PRL 30, 895 (1973); PRL 32, 527 (1974), Phys. Rev. A 10, 1277 (1974).

- Dependence of self-focusing of liquid crystal EBBA in isotropic state on the temperature (T_K = 78°) at pulse duration t_p = 15 ns. For CS_2 P_{cr} ~ 8 kW [2]

T [°C]	τ	t_p/τ	n_2 [10^{-11}esu]	P_{cr} [kW]
79.7	72.5	0.21	237	0.071
82.7	32.1	0.47	111	0.151
96.0	7.25	2.07	33.4	0.503
112.0	2.87	5.2	18.2	0.926
130.8	1.33	11.3	11.8	1.420

In Ref. 2, by variation of temperature self-focusing varied from transient [3,4] to quasi-steady-state.

[2]. Hanson, Shen, and Wong, Appl. Phys. 14, 65 (1977).
[3]. Akhmanov, Sukhorukov, Khokhlov, JETP 24, 198 (1966).
[4]. Fleck and Kelly, APL, 15, 313 (1969).

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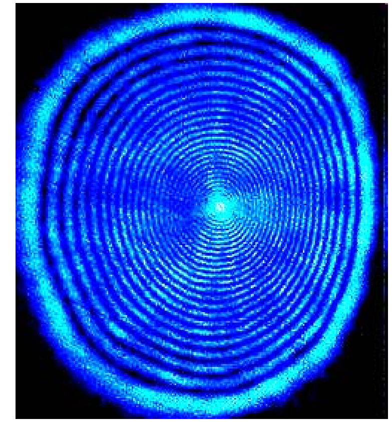
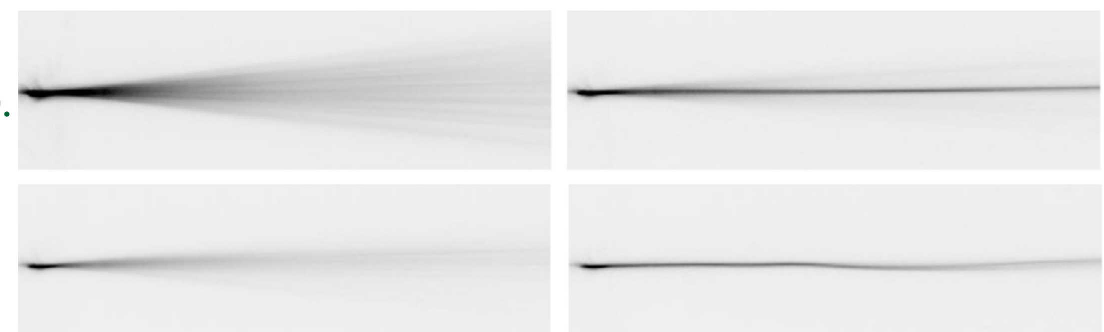
The giant optical orientational nonlinearity of nematic liquid crystal layers (nine orders of magnitude higher $\chi^{(3)}$ than for CS_2) [1]

Its response time is slow (submilliseconds to seconds).



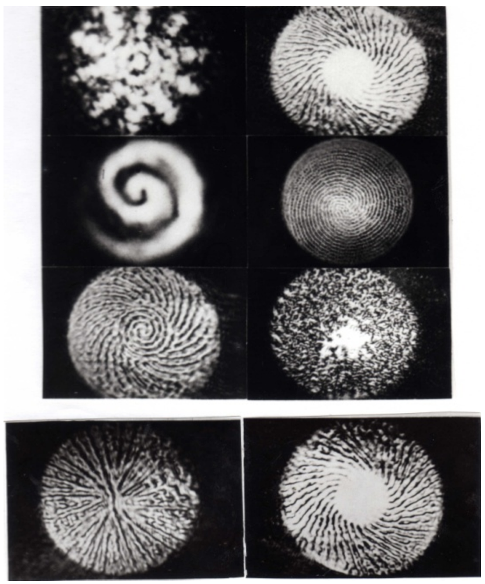
- Spatial self-phase modulation was observed under CW-irradiation in thin layers (~100μm) of aligned liquid crystals [2,3] as the result of optical Freedericksz transition (reorientation of liquid crystal molecules in the field of light wave);
- Adding a small amount of dye diminishes the threshold of Fredericksz transition on additional 1-2 orders of magnitude;
- Spatial solitons (nematicons) were observed (medium with saturation of nonlinearity) [4].



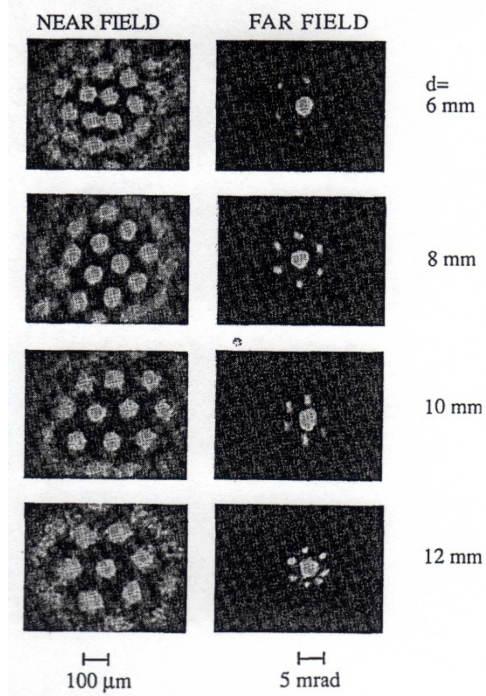
1. Zel'dovich, Pilipetskii, Sukhov, and Tabiryan, JETP Lett 31, 263 (1980).
2. Zolot'ko, Kitaeva, Kroo, Sobolev, Csillag, JETP Lett. 32, 158 (1980).
3. Durbin, Arakelyan, Shen, Opt. Lett. 7, 1145 (1982).
4. Assanto and Peccianti, IEEE J. Quant. Electr. 39, 13 (2003).

Optical pattern formation – "optical view" on nonlinear wave dynamics

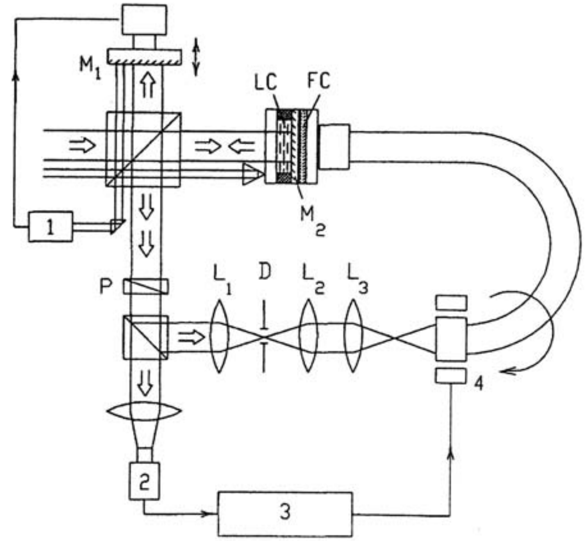
Liquid-crystal light valve, nonlinear interferometers, single feedback mirror with a thin liquid crystal layer

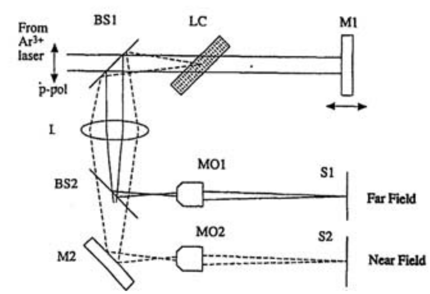


Akhmanov, Vorontsov, Ivanov, JETP Lett. 47, 707 (1988); JOSA B 9, 78 (1992)



Tamburini, Bonavita, Wabnitz, Santamato, Opt. Lett. 18, 855 (1993)





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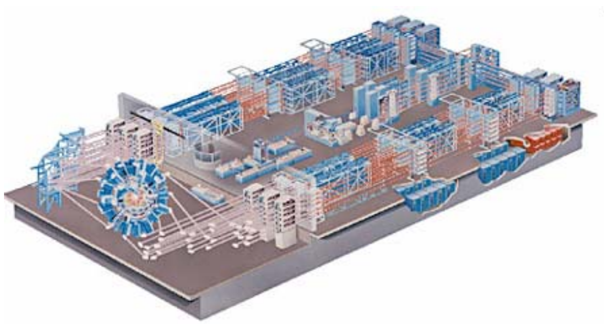
Applications include the construction of optical neural networks, pattern recognition and associative memories.

The 60 beam OMEGA laser at the University of Rochester has 365 liquid crystal optical elements



There are two liquid crystal component types with apertures of 100 mm to 200 mm:

- cholesteric circular polarizers;
- nematic waveplates

High laser damage resistance >9 J/cm^2 @1054 nm / 1 ns

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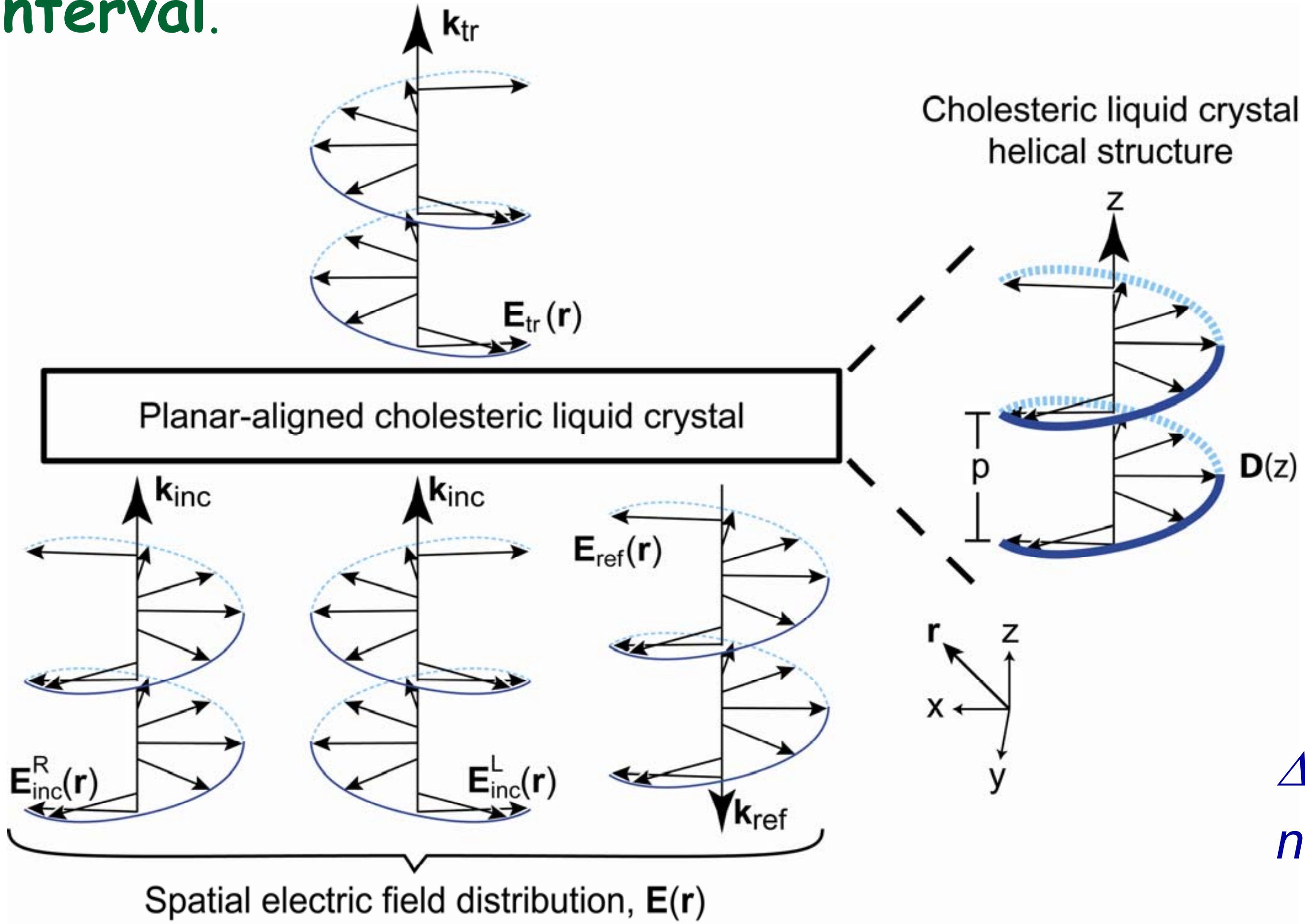
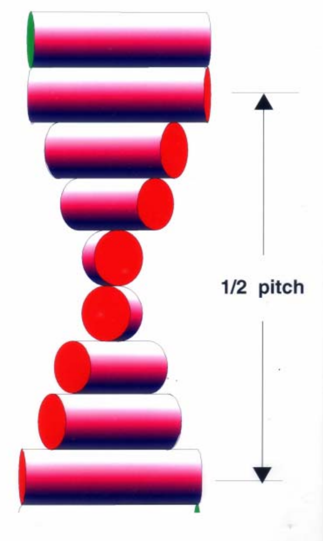
Athermal Cholesteric Liquid Crystal Pitch Dilation by the Field of a Light Wave

Planar-aligned cholesteric layer is a photonic bandgap structure which acts as a mirror for circularly polarized light of definite handedness in some spectral interval.





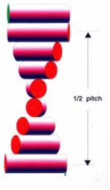
$$\lambda_c = n_{av}\, p$$

$$\Delta\lambda = p(n_e - n_o)$$

$$n_{av} = (n_e + n_o)/2$$

In 1982 Winful calculated that athermal, light-induced changes in the pitch of the cholesteric helix lead to a reflectivity drop and bistable reflection characteristic from a cholesteric mirror

For helix-pitch changes light intensities should be larger than ~MW/cm^2



Winful, PRL, **49**, 1179 (1982).

- **At short-pulse intensities exceeding by an order of magnitude the estimated value cholesteric mirror reflectivity drop was not observed.**

- **Laser pulse duration at ~MW/cm^2 should be of the order of helix unwinding time ~several milliseconds!**

H. Espinet et al., J. Appl. Phys. **59**, 1386 (1986).

- **In 1987, Lee et al. demonstrated that a CW laser may induce subtle phase changes in a resonance cholesteric structure.**
- **This was interpreted as manifestation of Winful's prediction of field-induced pitch dilation.**

Lee, Jacobs, Gunderman, Schmid, et al., Opt.Lett. **15**, 959 (1990), Mol. Cryst. Liq. Cryst. 150b, 617 (1987).

Issues left unanswered:

- Winful's other prediction of a drop in cholesteric layer reflectivity was not observed.
- What happens in much more intense fields of pulsed lasers?
- How to distinguish a laser-induced pitch dilation in cholesteric helices from a thermal nonlinearity?

In our experiments:

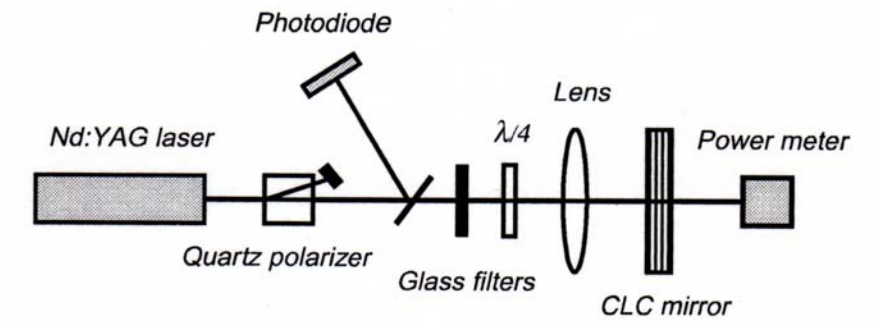
- **Reflectivity drop of cholesteric mirrors was observed using pulse repetition rate laser with an accumulation effect from many pulses.**
- **To distinguish a field-induced orientational effect from thermal changes of a cholesteric pitch, the laser was switched to CW-operation with the same average power density as in a pulsed regime.**

Lukishova, Lebedev, Magulariya, Belyaev, Malimonenko, Schmid, Bull. Russ. Acad. Sci., Phys. **59**, 2086 (1995);
JETP Letts. **63**, 423 (1996),
Quant. Electron, 26, 796 (1996).

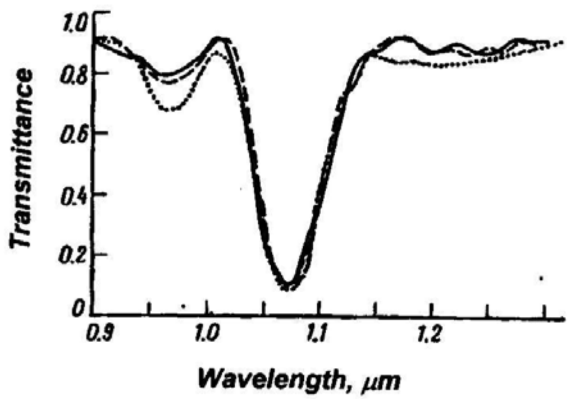
See also Grebe, Macdonald, Eichler, Mol. Cryst. Liq. Cryst., **282**, 309 (1996) with intracavity experiments at ~GW/cm^2 intensities.

1.06 μm laser operated at two modes: (1) CW; (2) 4.5 kHz repetition rate, 500 ns pulse duration

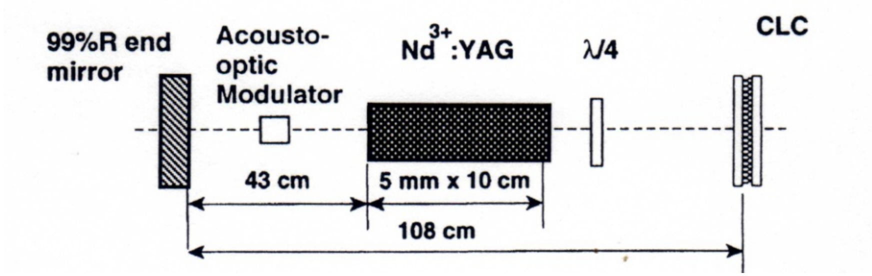
Free-space set up with a cholesteric mirror



Cholesteric mirror transmittance



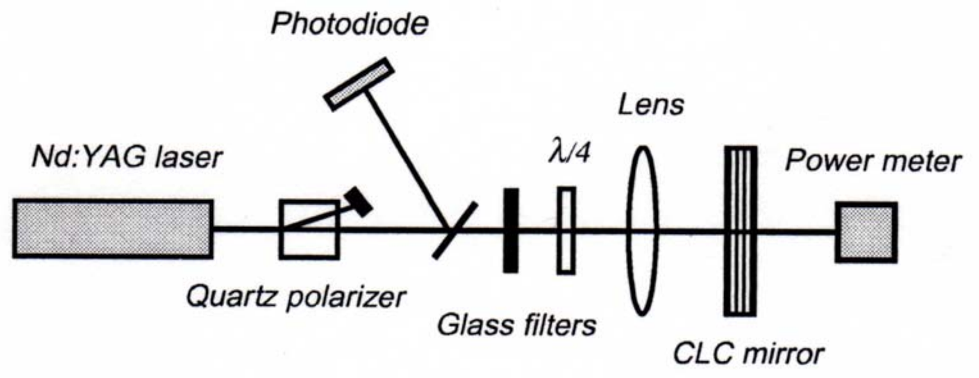
Laser resonator with a cholesteric mirror

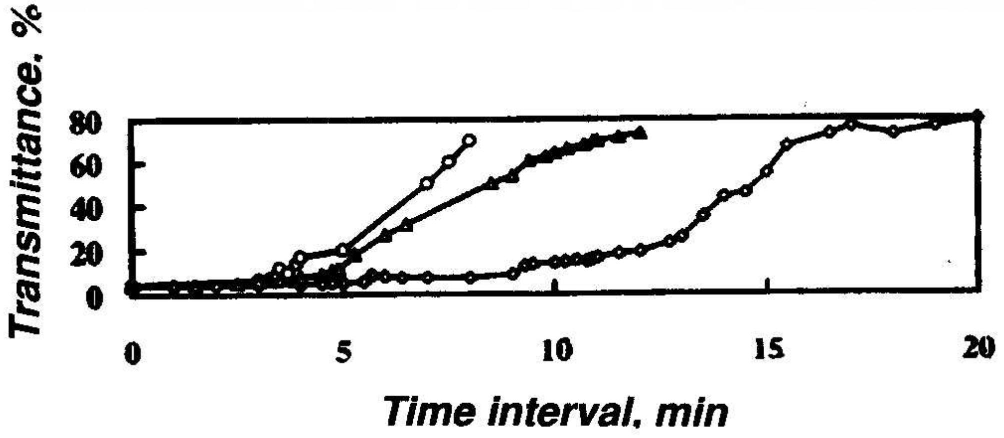


One inner side of each of 4 mirrors had a strong anchoring Opposite site had a weak anchoring.

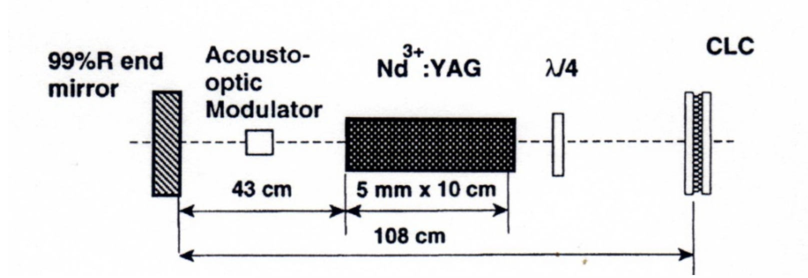
Nonlinear selective reflection experiment

1.06 μm, 4.5 kHz, 500 ns, 1-10 MW/cm²

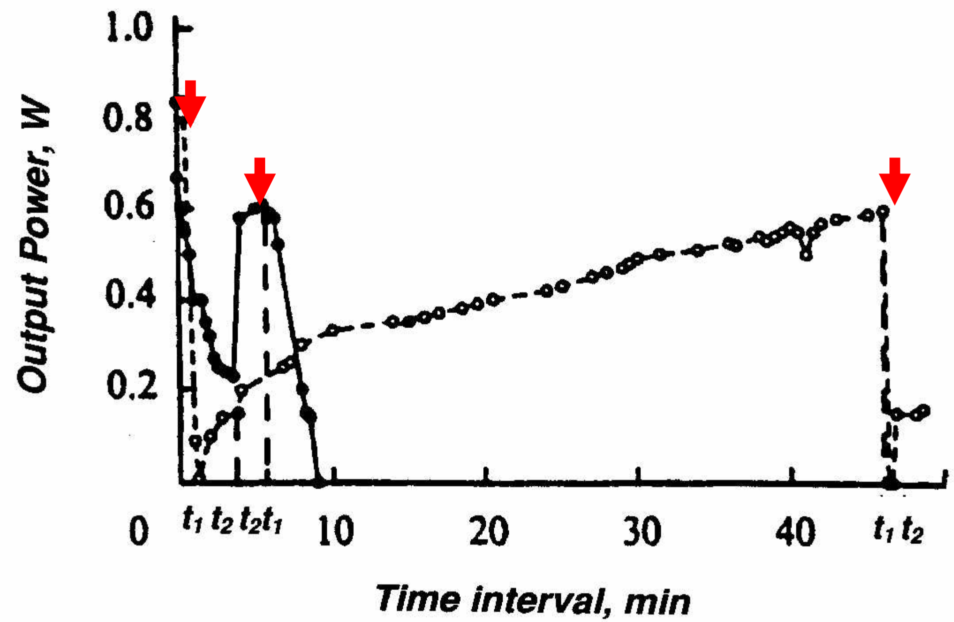




Effect of reflectivity drop disappears at CW-operation (effect does NOT depend on AVERAGE power density)



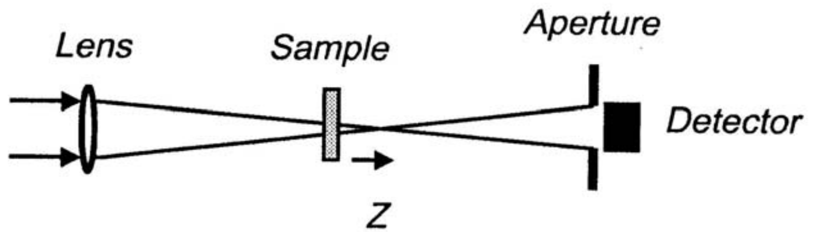
At 4.5-kHz, 500-ns, the cholesteric mirror reflectivity drops and lasing ceases, switching to a CW-mode recovers lasing.

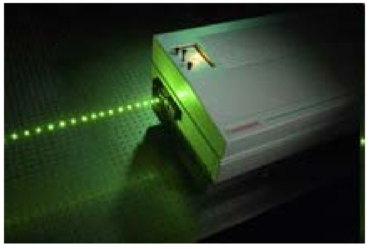


Effect was not observed if the weak-anchoring side of the mirror was its entrance side (flipping of the mirror on 180°)

Nonlinear Absorption and Refraction of Liquid Crystals under Nanosecond Laser Irradiation

Strong nonlinear absorption of thermotropic liquid crystals was observed in optical power limiting studies and z-scan measurements under visible-light, pulsed laser irradiation





- For the nematic phase, the two-photon absorption coefficient has a several-times higher value for incident polarization parallel to the molecular dipole direction than for perpendicular polarization [1].
- Heating of thermotropic liquid crystals by short-pulse laser irradiation in the visible range that drives photoacoustical and thermotropic effects, is caused by two-photon absorption, concurrent or subsequent excited state absorption, and the efficient decay of the excited states through radiationless-recombination channels [2, 3].

[1] Durbin and Shen, Phys Rev A30, 1419(1984).

[2] Deeg and Fayer, J. Chem. Phys. 91, 2269 (1989).

[3] Macdonald and Eichler, Appl. Phys. B60, 543 (1995).

Nonlinear refraction of liquid crystals under high-power, nanosecond laser irradiation in the presence of two-photon absorption

In addition to the orientational nonlinearity, refractive index changes can be caused by heating

$$\Delta n = (\partial n / \partial \rho)_T \Delta\rho + (\partial n / \partial T)_\rho \Delta T$$

Thermal-density nonlinearity — Build up time $t_{ac} = r_o/V_S$

Thermooptical effects (thermal lens) — Build up time of tens-hundreds of nanoseconds

- For isotropic liquid crystals at the several-nanosecond time scale and several tens-micrometers beam-waist diameter, transient molecular-reorientation and thermal/density refractive nonlinearities compete in changing the sign of the total transient refractive nonlinearity.
- For planar-aligned nematic liquid crystal layers, cumulative effects in heating (and refractive nonlinearity) were observed even at low, 2-10 Hz pulse repetition rate as the result of strong two-photon absorption for planar-aligned nematic layera and extraordinary large thermooptics coefficients, e.g., for nematic 5CB $\partial n_{par} / \partial T \sim -2.5 \cdot 10^{-3} \text{grad}^{-1}$.

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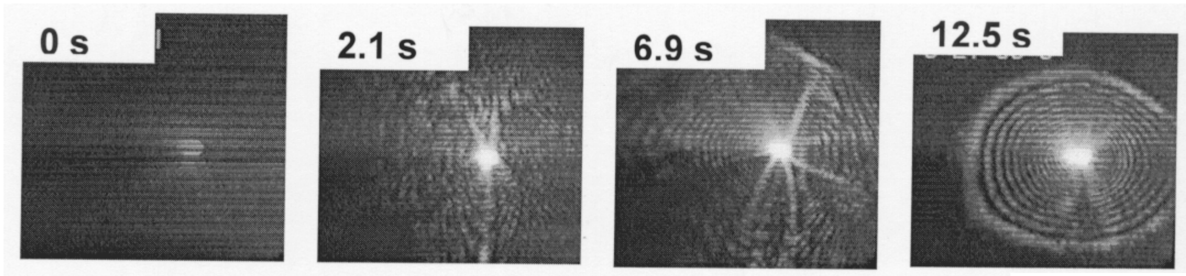
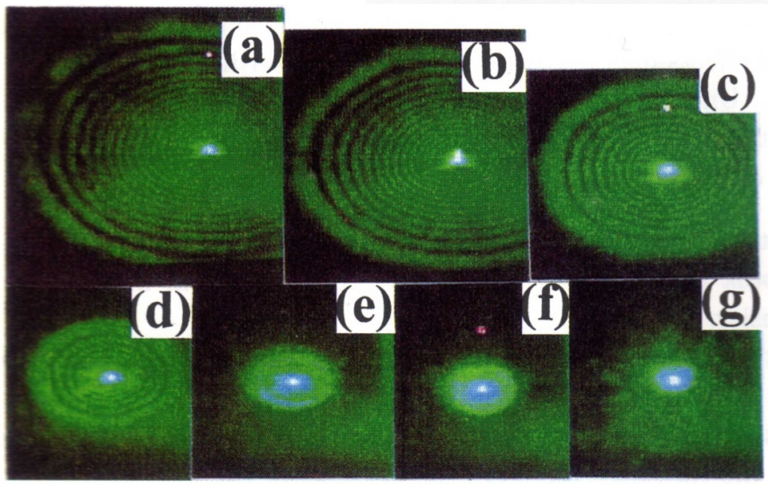
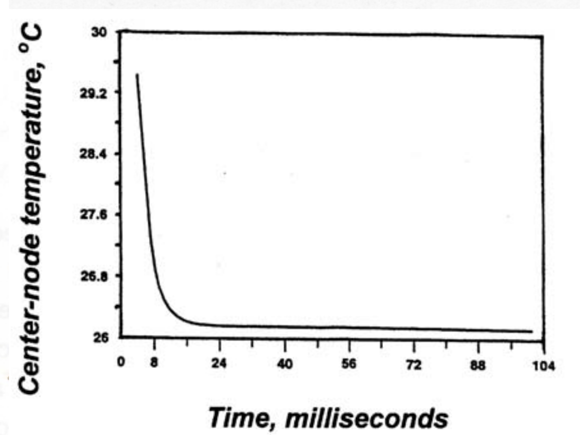
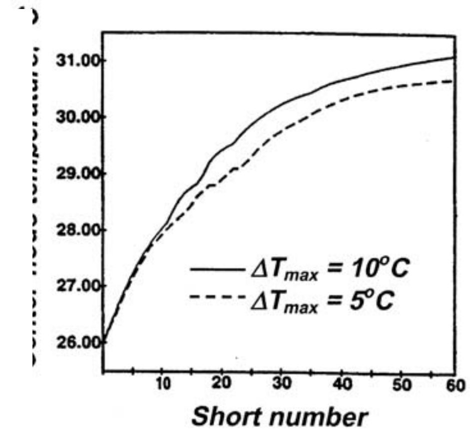
Cumulative effects in heating of planar-aligned LC-cells at 0.5 GW/cm^2, 532-nm, 6-ns pulse duration, 10Hz pulse repetition rate, 125-μm thickness cell, 160-μm beam diameter





Changing the thermal self-phase modulation rings with diminishing incident intensity from (a) to (g).





Numerical modeling of heat diffusion (ANSYS/Thermal code): Left: center-node temperature relaxation in time after the first pulse; Right center-node temperature versus the number of pulses.

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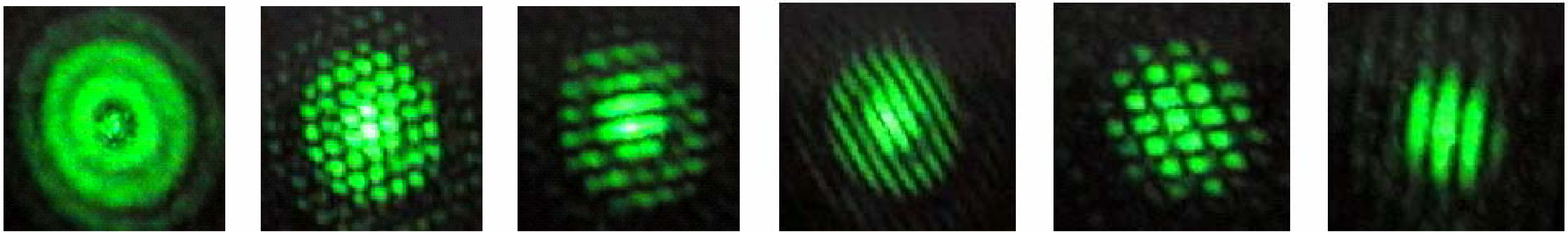
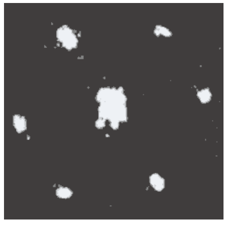
S.G. Lukishova, J. Nonl. Opt.Phys.& Mater. 9,365 (2000).

Feedback-free Pattern Formation in Dye-doped Liquid Crystals and Isotropic Liquids

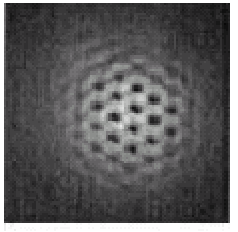


Generally optical feedback is necessary for hexagonal pattern formation in nonlinear optics

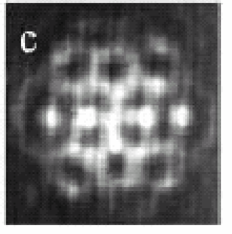
1. M.A. Vorontsov and W.B. Miller, Eds., *Self-Organization in Optical Systems and Applications in Information Technology,* Springer (1985).
2. Transverse Effects in Nonlinear Optical Systems, Special issues of *J. Opt. Soc. Am*. B7, is. 6 and 7 (1990) with overview of N.B. Abraham and W.J. Firth.
3. S. Residori, *Physics Reports* 416, 201-272 (2005).



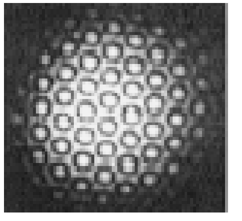
Grynberg et al. (1986)



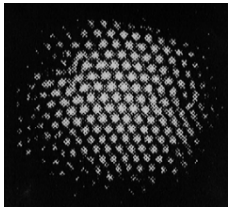
Ackerman et al.(1995)



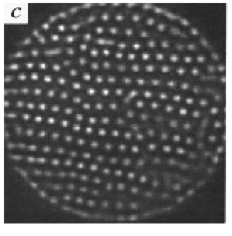
Vaupel et al. (1999)



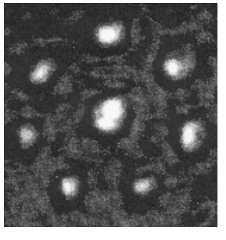
Luchnikov et al. (1999)



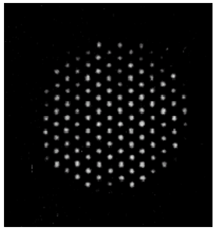
Banerjee et al. (1995)



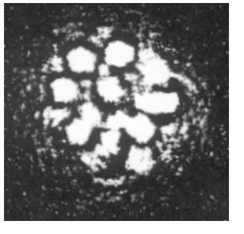
Vorontsov et al. (2000)



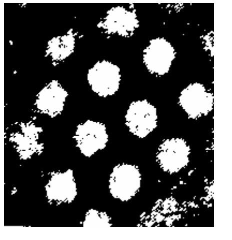
Arecchi et al. (1994)



Neubecker et al. (1995)

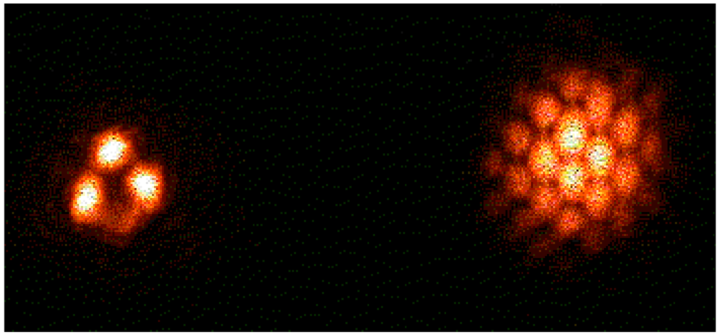


Macdonald et al. (1992)

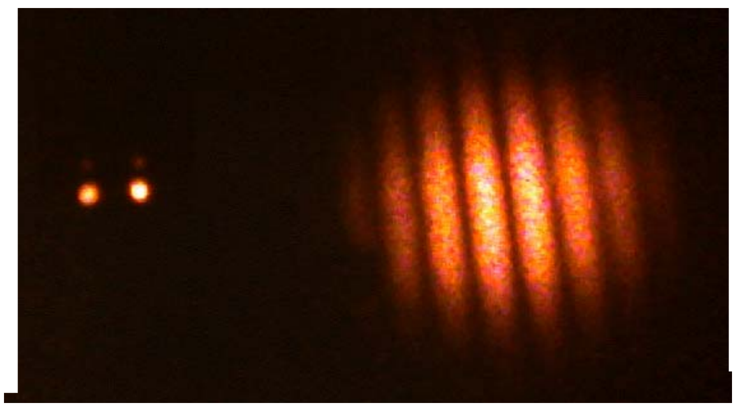


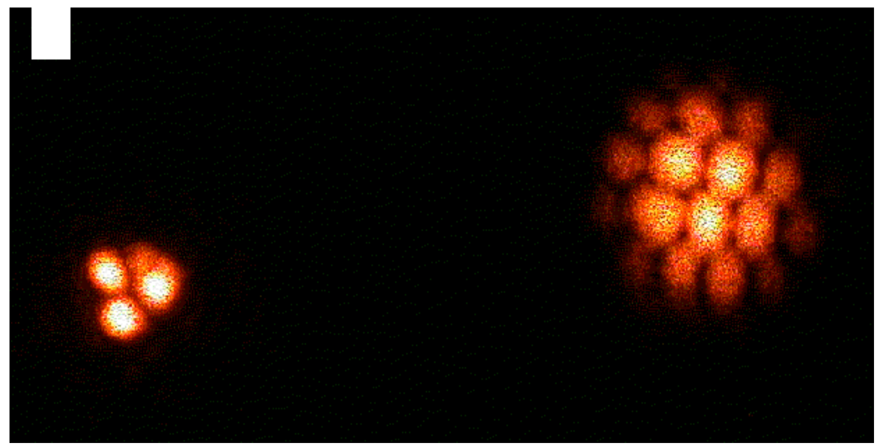
Tamburrini et al. (1993)

Hexagonal pattern formation in a feedback-free nonlinear optical system



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





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

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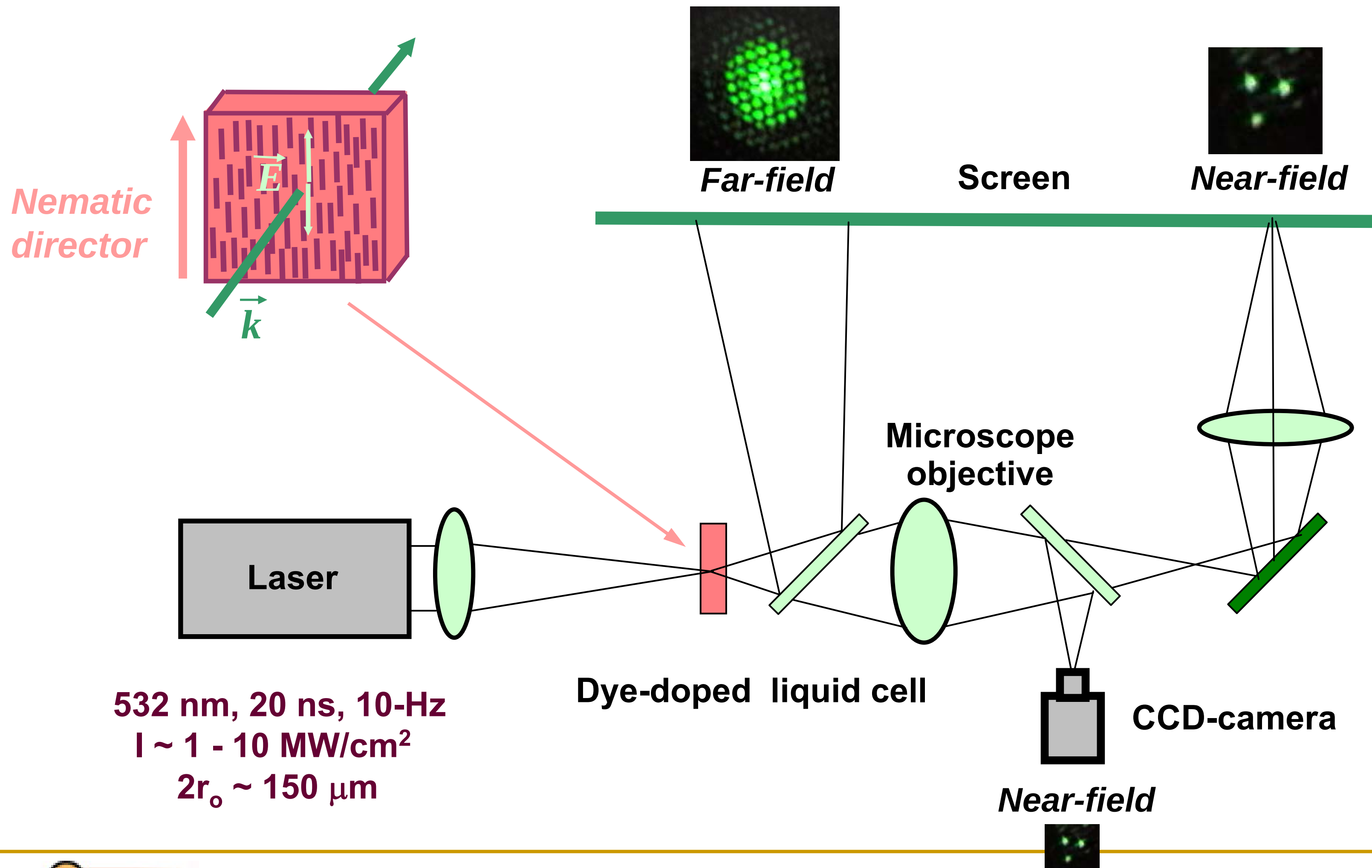
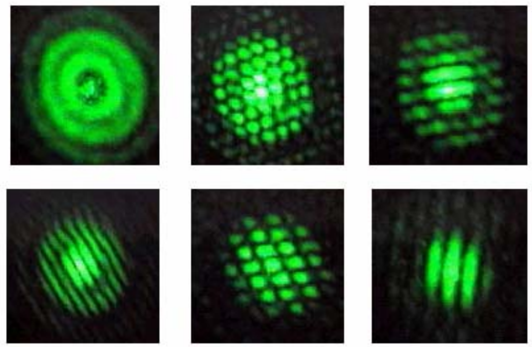
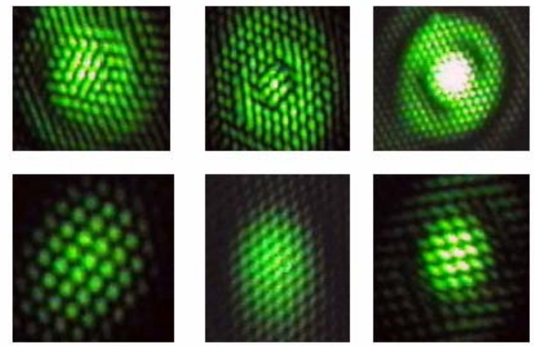
Experimental set up



Feedback-free kaleidoscope of patterns: far-field



50-cm from the output of planar-aligned E7 cell



50-cm from the output of unaligned E7 cell

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;

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

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

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

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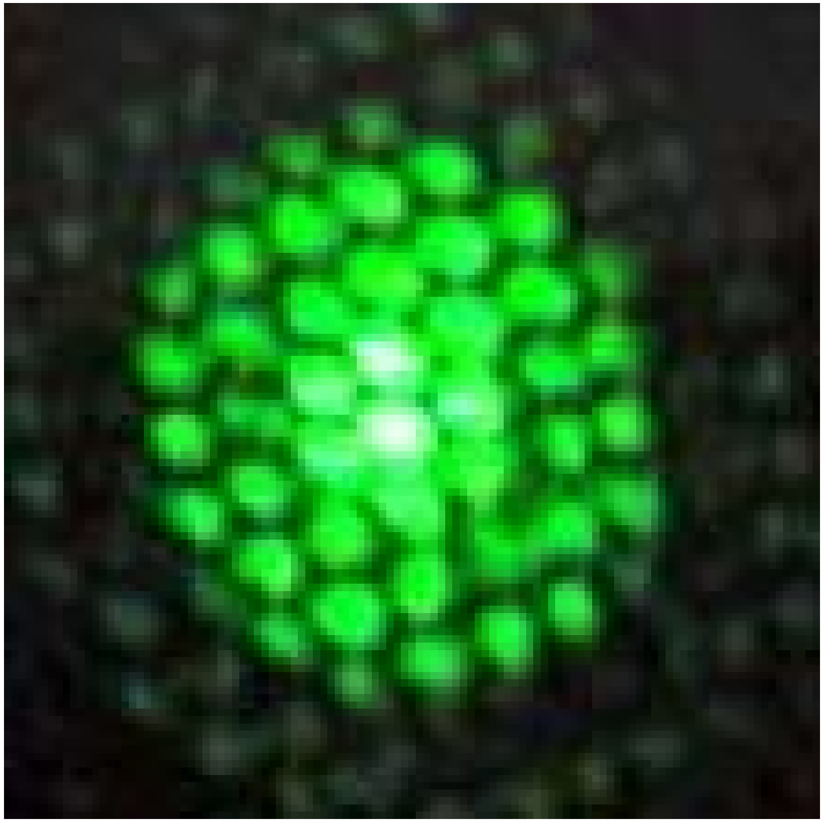
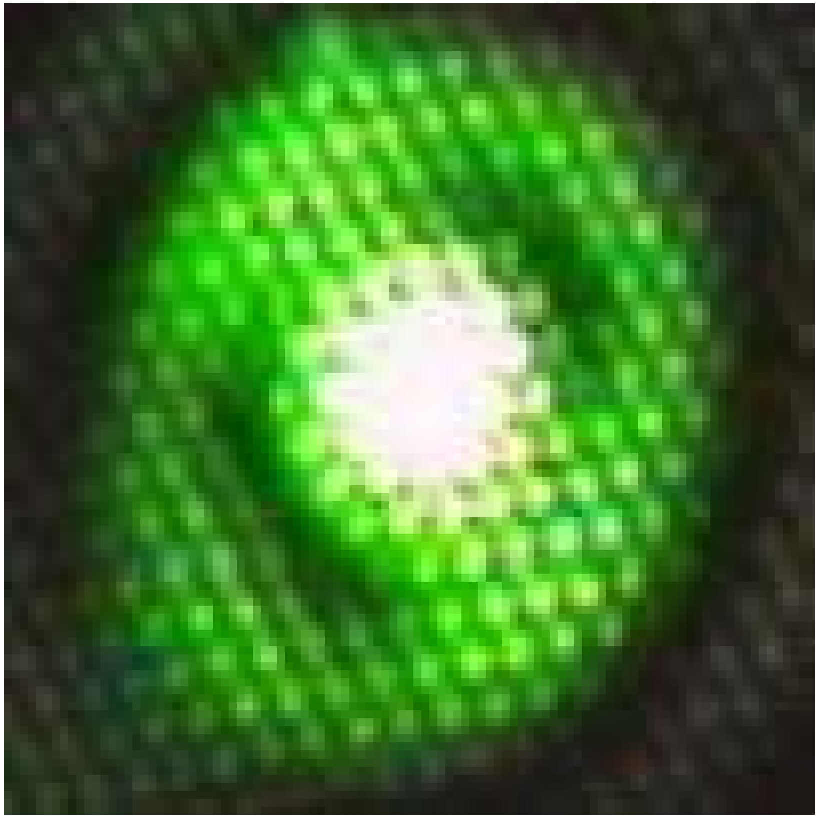
Hexagons in the far-field

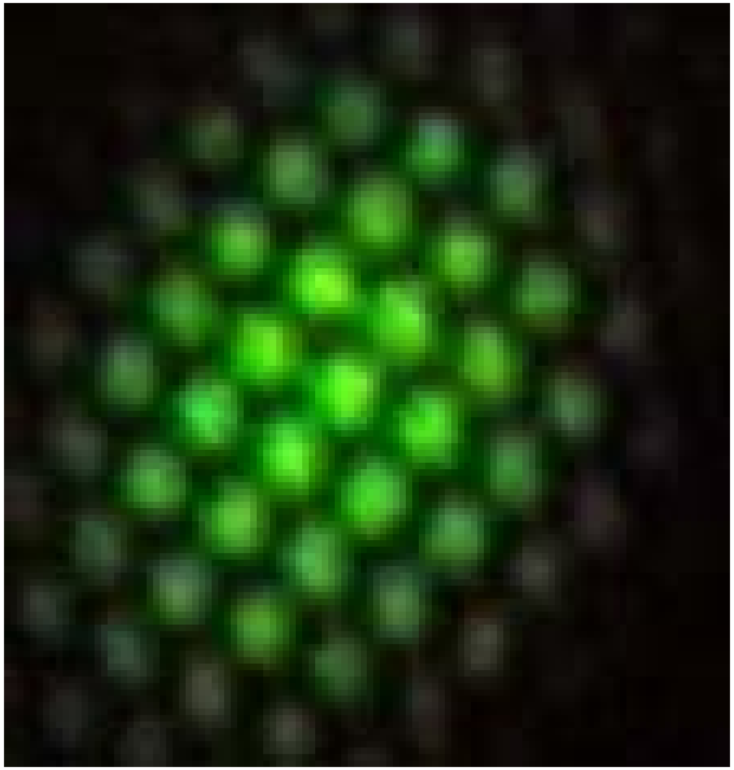
I ~ 1 – 10 MW/cm^2, 20 ns, 10 Hz prr)



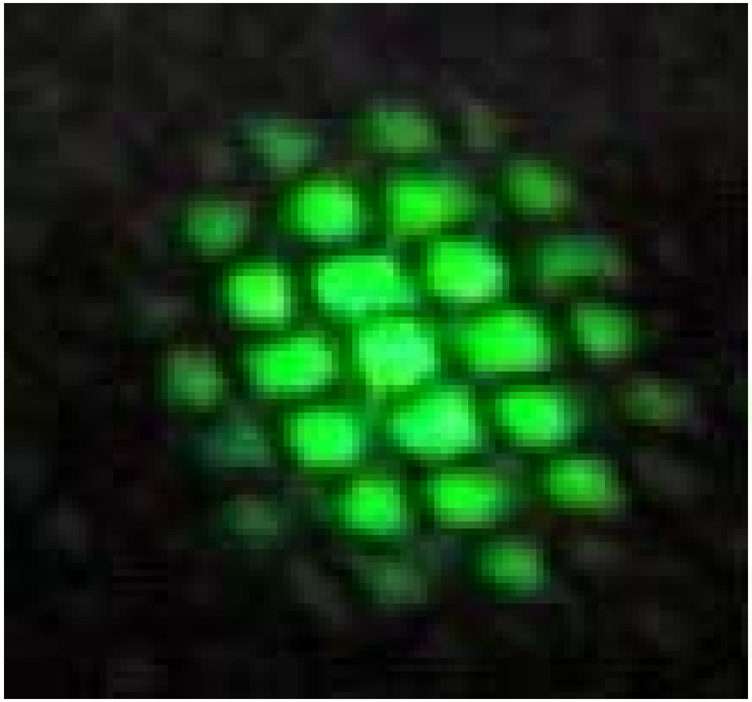
Hexagons in the far-field (continued)



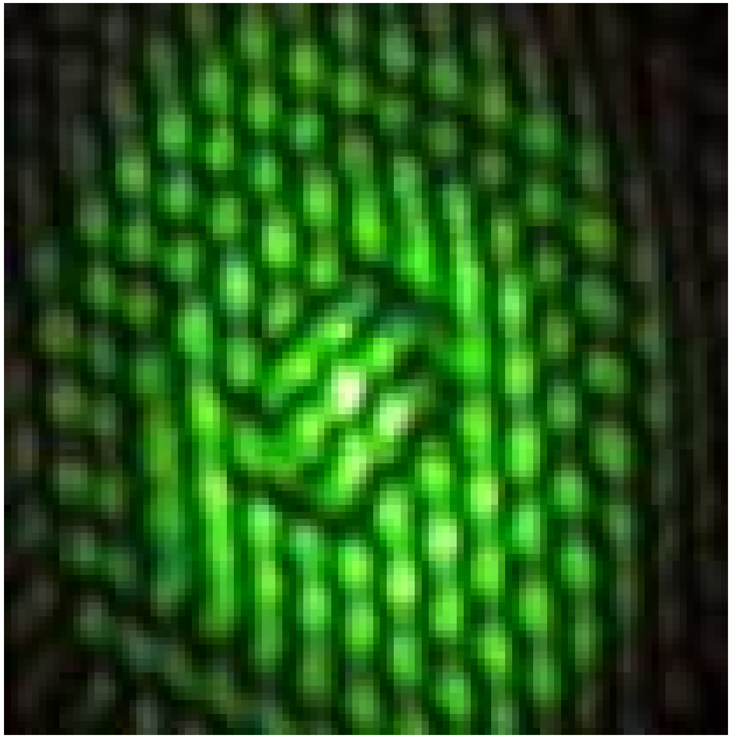
Hexagons in the far-field (continued)



Hexagons in the far-field (continued)



Hexagons in the far-field (continued)



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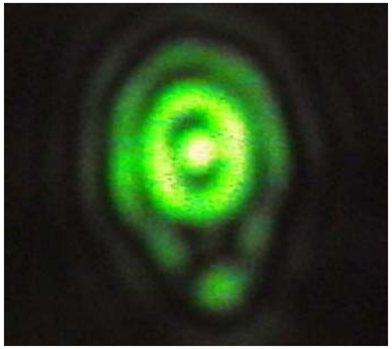
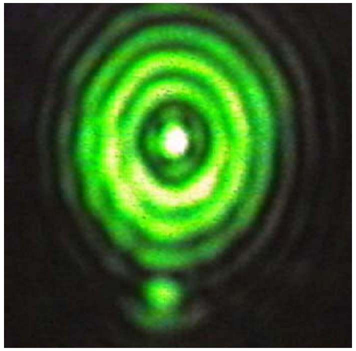
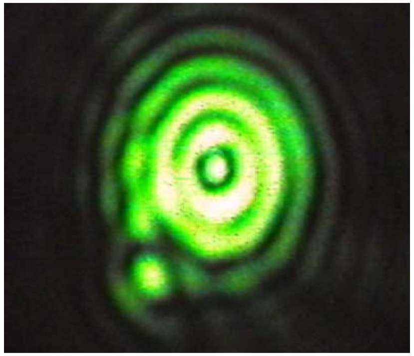
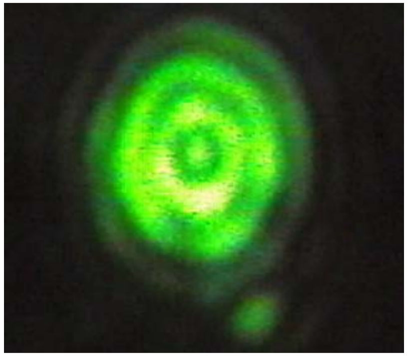
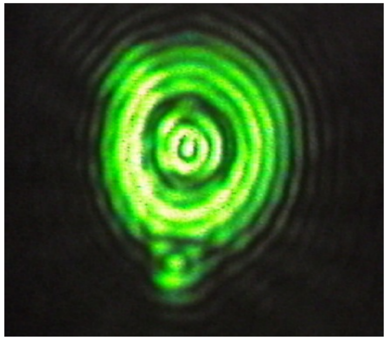
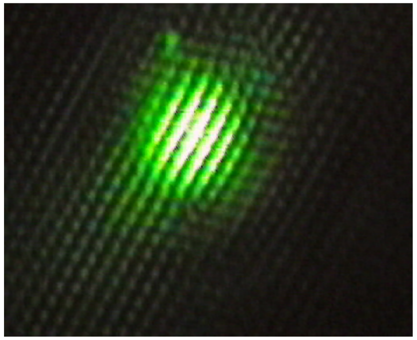
Localized structures in pattern formation in unaligned liquid crystals

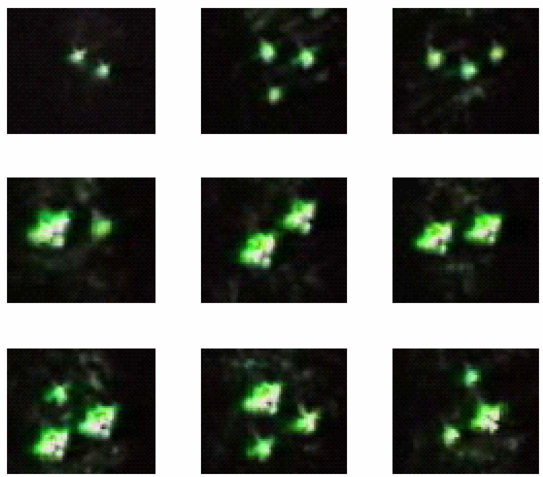








Feedback-free kaleidoscope of patterns: near-field

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

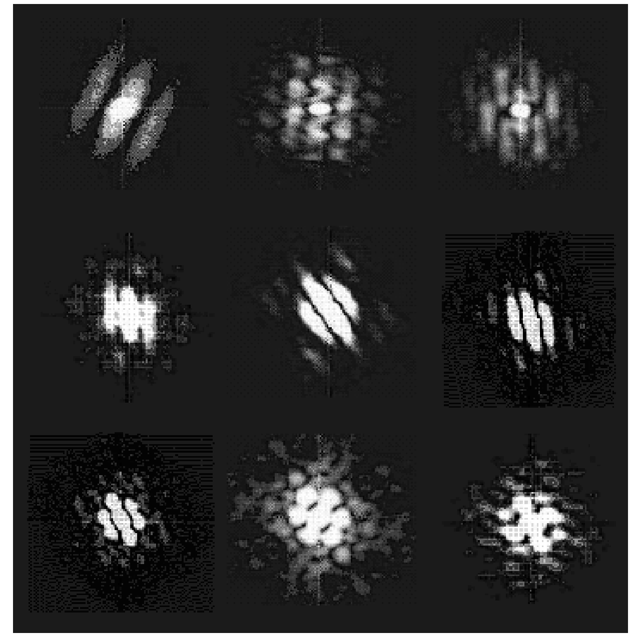
140μm



500 x optical-system-magnification

The size of the spots d_a ~ 5 - 15 μm with distance between spots d_o ~ 35 - 70 μm.

(Calculated from the far-field experiments d_a = 5 - 16 μm; d_o = 32 - 81 μm).



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

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

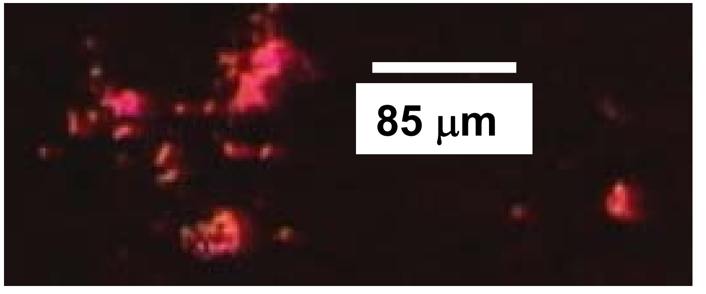
- Diffraction of the incident laser beam on the one/two/three/or more several-micron-size "drops" with absorption and/or refraction properties different from the surrounding material.
- The patterns' ring structure can be attributed to the diffraction of laser light at the sharp edge of "drops".
- The 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.

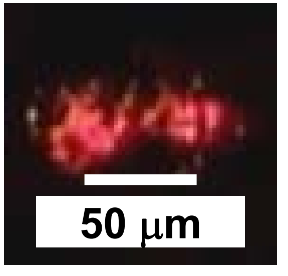
Possible mechanism of "drop" formation

- Phase separation in the multicomponent solution at elevated temperature/temperature gradient (e.g., Soret effect (diffusion of the dye molecules in the temperature-gradient-field)).

Optical microscope images of the cells after laser irradiation show dye adsorption to a substrate

5CB liquid crystal cell between cross polarizers





Light-induced phase separation of the dye molecules from the liquid crystal host and adsorption to a substrate of the cell were observed also by D. Voloschenko and O.D. Lavrentovich (J. Appl.Phys., 86, 4843, 1999).

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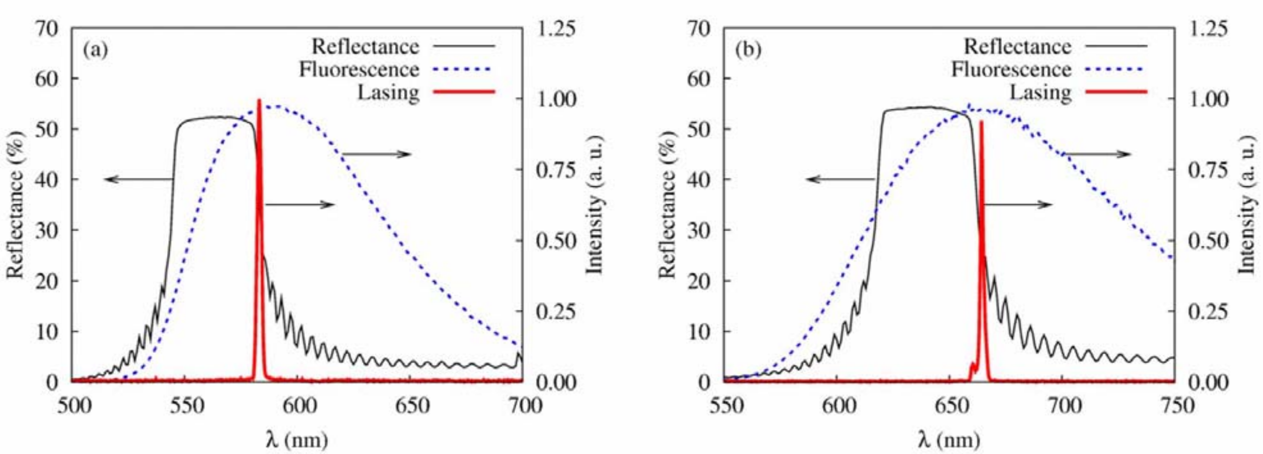
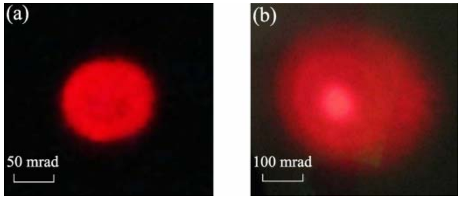
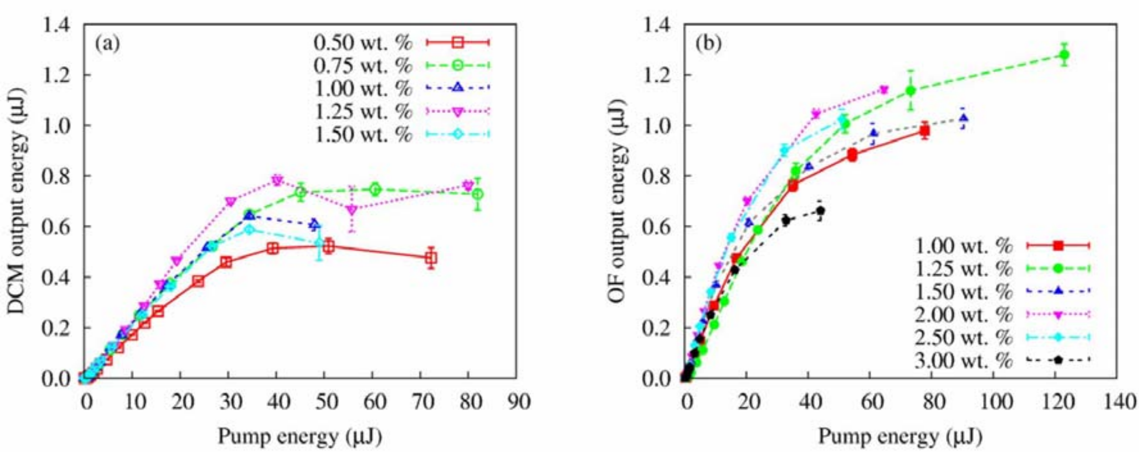
Cholesteric Liquid Crystal Lasers

1. I. Il'chishin, E. Tikhonov, V. Tishchenko, and M. Shpak, JETP Lett. **32,**24–27 (1978).
2. V.I. Kopp, B. Fan, H.K.M. Vithana, and A.Z. Genack, Opt. Lett. **23,** 1707–1709 (1998).
3. Review: H. Coles and S. Morris, Nature Photonics **4**, 678 (2010).
4. L.M. Blinov and R. Bartolino, Eds, *Liquid Crystal Microlasers* (2010).

Cholesteric laser based on monomeric (fluid-like) structures with an oligofluorene dopant.

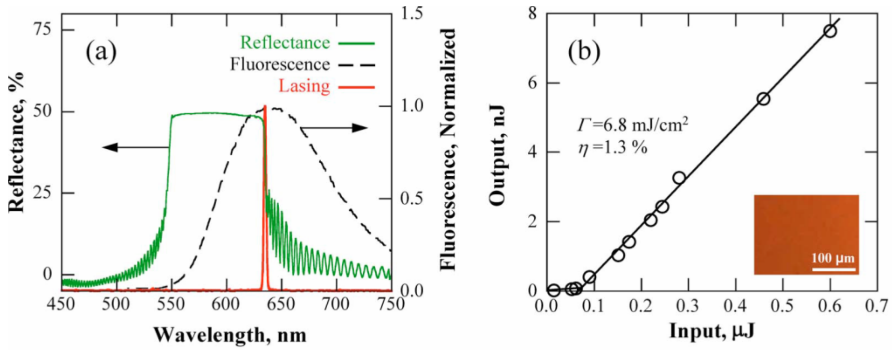


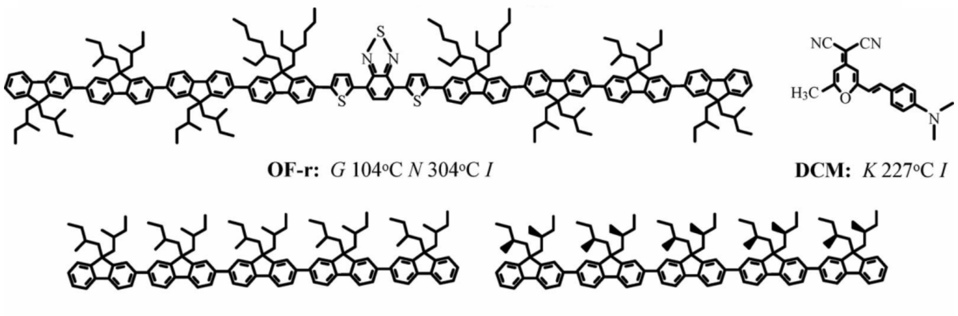




K. Dolgaleva, S. K. H. Wei, S.G. Lukishova et al., JOSA B **25**, 1496 (2008).

Glassy oligomeric (solid) cholesteric liquid crystal lasers doped with oligofluorene



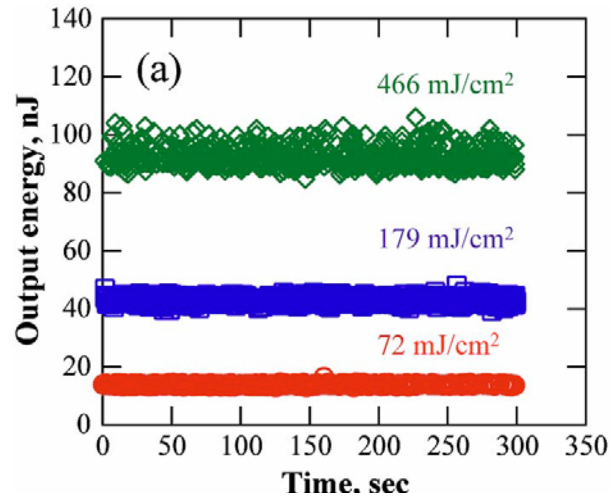


OF-r: *G* 104°C *N* 304°C *I*

DCM: *K* 227°C *I*

F(MB)5-N: *G* 92°C *N* 171°C *I*

F(MB)5-Ch: *G* 91°C *Ch* 173°C *I*



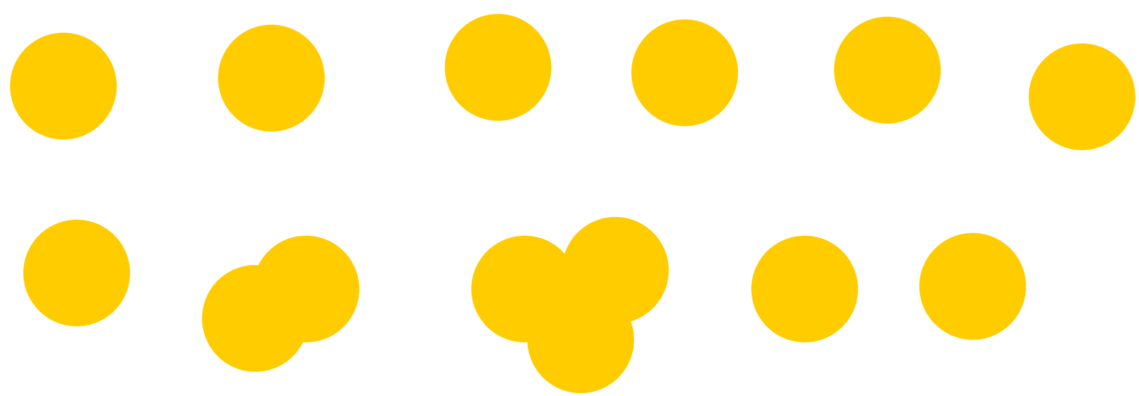
S.K.H. Wei, S.H. Chen, K. Dolgaleva, S.G. Lukishova, and R.W. Boyd, APL **94**, 041111 2009.

Single-Photon Sources Based on Single-Emitter Fluorescence in Liquid Crystal Hosts

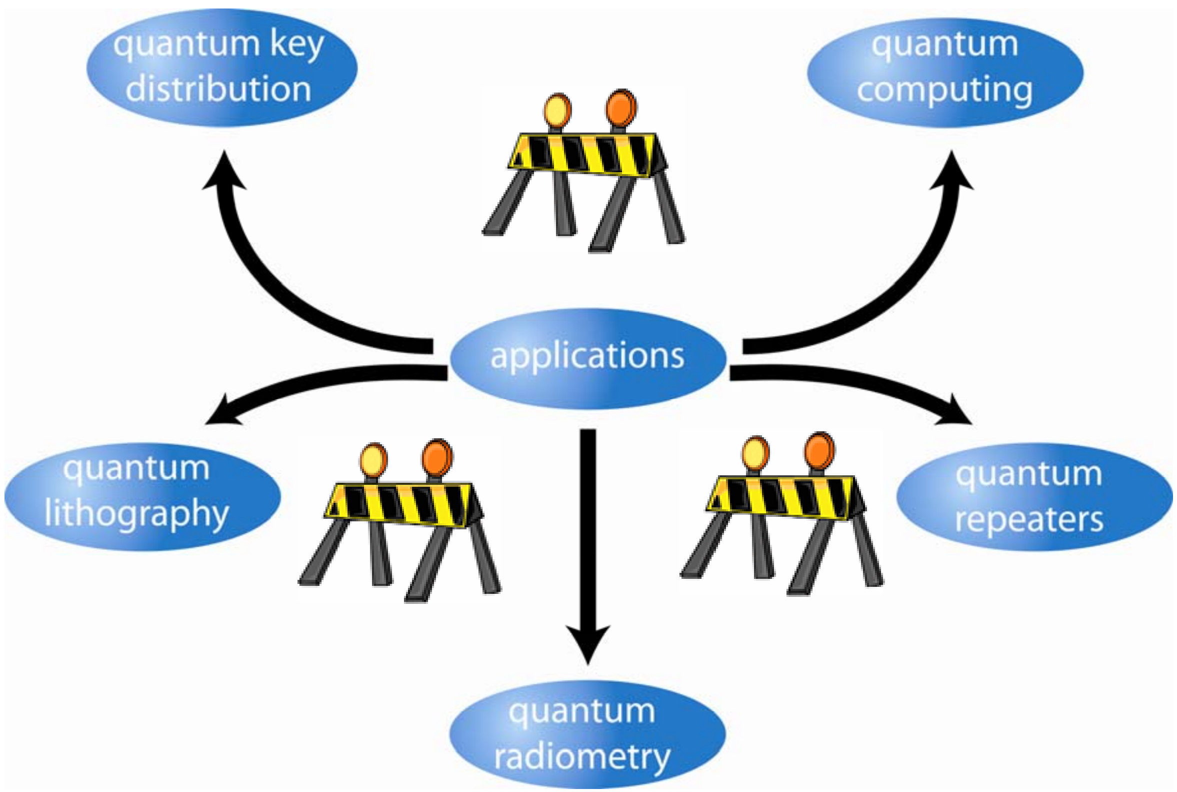
Single-Photon Sources produce photons exhibiting antibunching (separation of all photons in time)

Single-photon source

Laser light attenuated to a single-photon level



Applications of single-photon sources



To increase the efficiency of quantum cryptography system based on BB84 protocol single photons should have definite polarization

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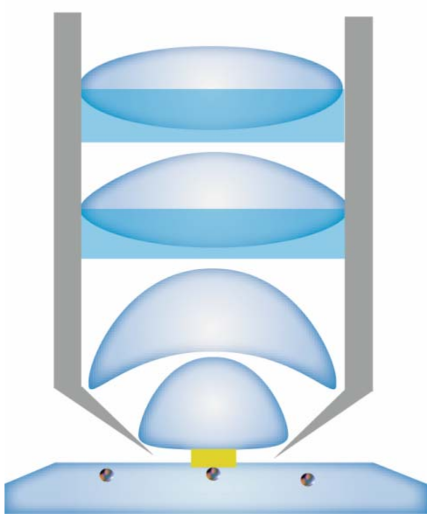
How do we realize a SPS?

To produce single photons, we start with a sample area containing a very low concentration of emitters.

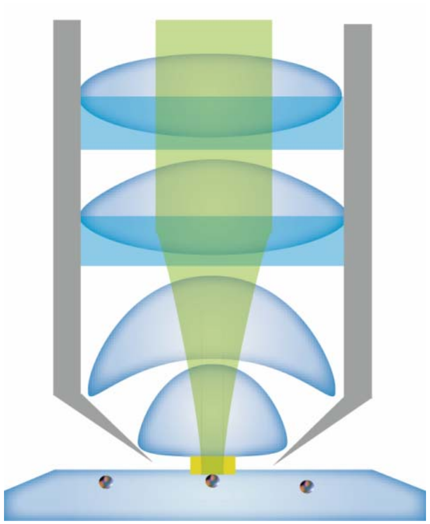
microscope objective →



How do we realize a SPS?

A laser beam is tightly focused onto the sample, so that only one emitter becomes excited.

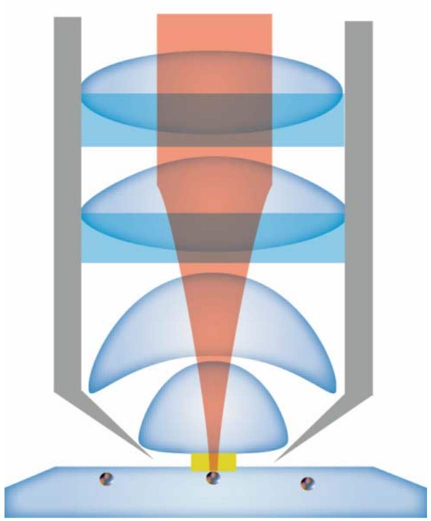
microscope objective



How do we realize a SPS?

The emitter fluoresces only one photon before subsequent optical excitation, thus realizing a single-photon source.

microscope objective →



To enhance single photon efficiency a cavity should be used

Sample preparation

Emitters:

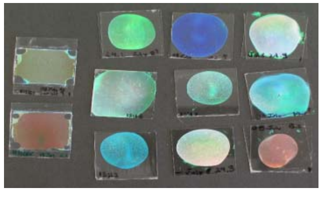
- colloidal semiconductor quantum dots (CdSe/CdS, CdSeTe and PbSe);
- single dye molecules (DiI and terrylene).
- single color centers (NV) in nanodiamonds

Hosts [liquid crystals in monomeric (fluid) or oligomeric (solid) form]:

- cholesteric (1-D chiral photonic bandgap structure);
- nematic (align molecular dipoles in one direction)

Fabrication method

Liquid crystal structures were fabricated by planar alignment of the mixtures (using buffing, photoalignment or shearing two substrates relative to each other)



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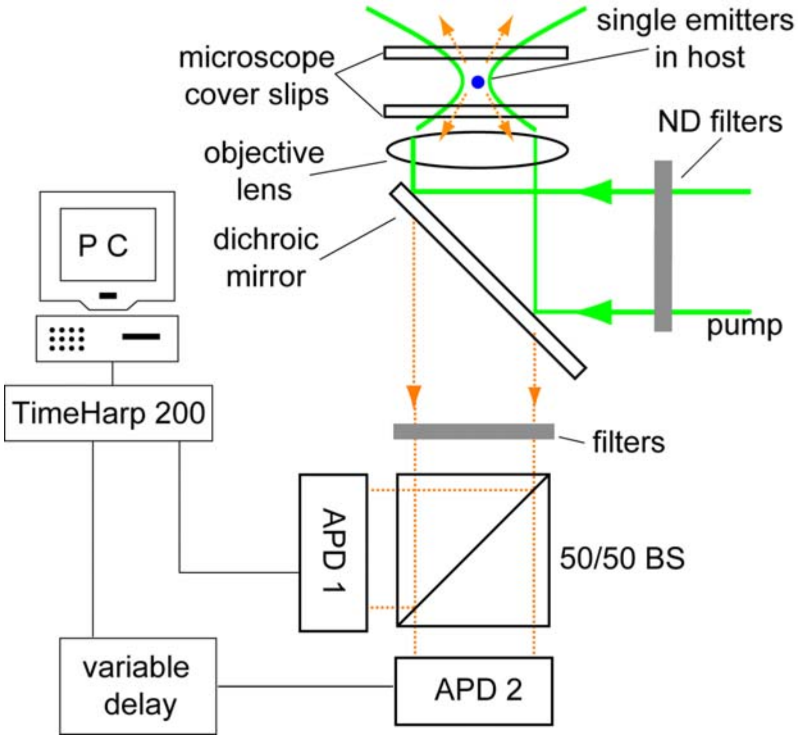
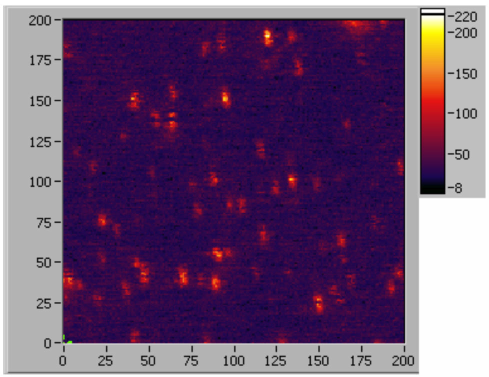
S.G. Lukishova et al., *IEEE J. Select. Top. in Quant. Electronics*, Special Issue on Quantum Internet Technologies, **9**, is.. 6, 1512 (2003).

Confocal fluorescence microscope and Hanbury Brown and Twiss setup
(photon antibunching measurements)

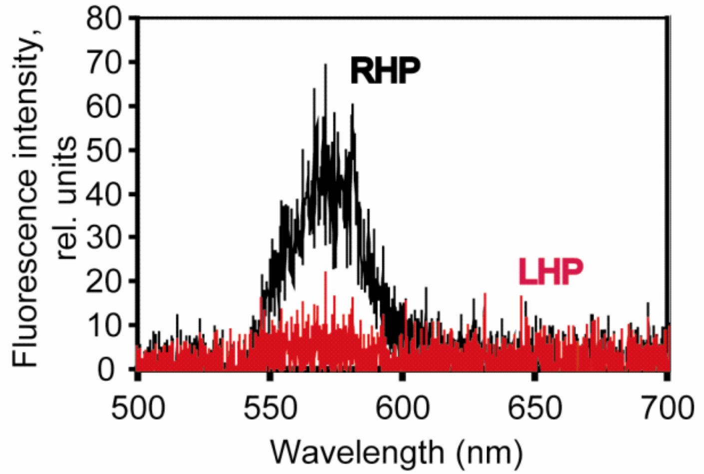


76 MHz repetition rate, ~6 ps pulsed-laser excitation at 532 nm, cw 514 nm argon ion laser, cw 975 nm diode laser, and cw 633 nm He-Ne laser.

Circularly polarized fluorescence from several single quantum dots in a photonic bandgap CLC host (left) and a fluorescence spectrum of the host (right)



15 μm x 15 μm scan



The degree of circular polarization is measured by the dissymmetry factor $g_e = 2(I_L - I_R)/(I_L + I_R)$ [1]. At 575 nm g_e = -1.6. For unpolarized light $g_e = 0$; for circular polarized light $|g_e| = 2$.

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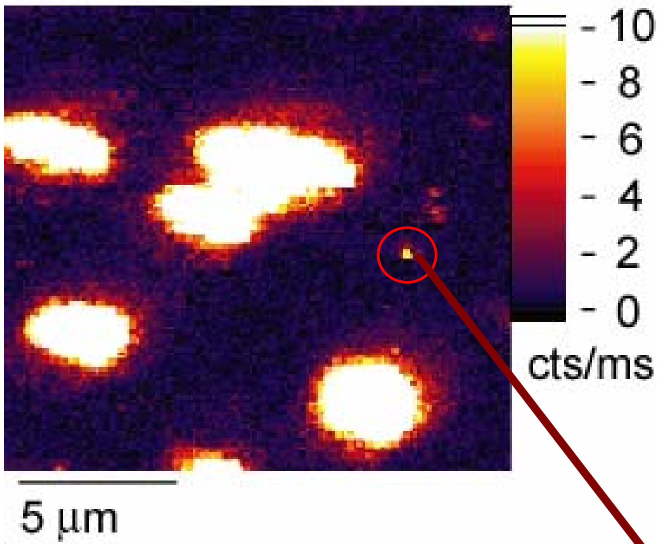
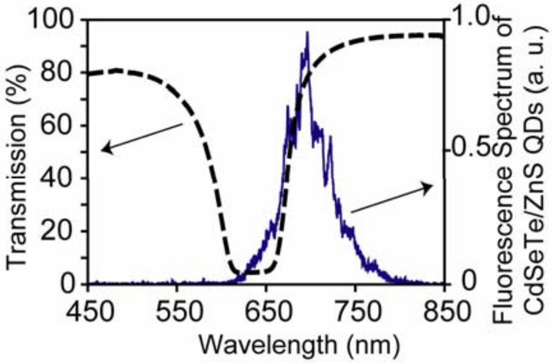
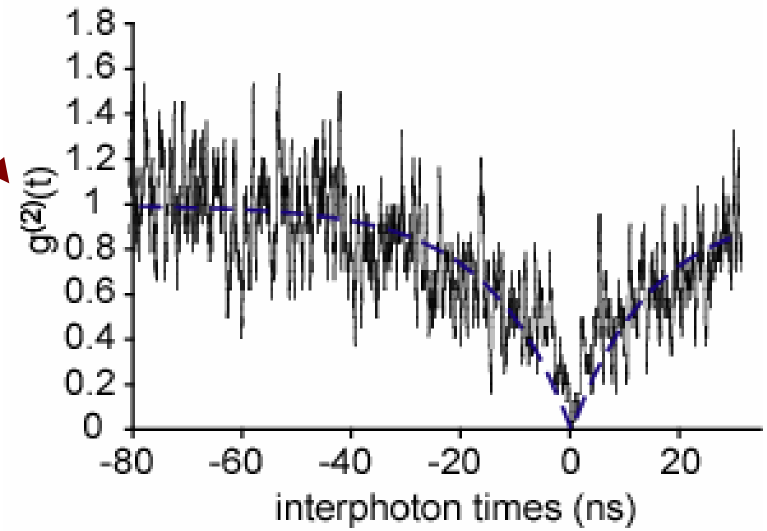
[1] S. H. Chen et. al, *Nature*, **397**, 506-508 (1999).

Fluorescence antibunching from a CdSeTe quantum dot in a 1-D photonic bandgap cholesteric host
$g^{(2)}(0) \sim 0$



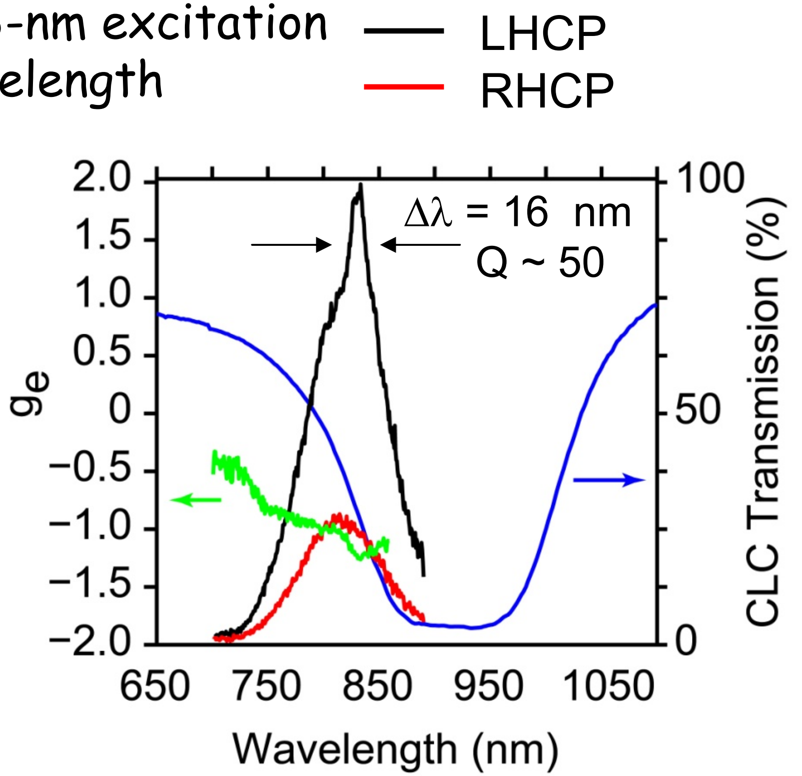
$\lambda_o = 705$ nm





Circular polarized microcavity resonance in photonic bandgap glassy in photonic bandgap glassy CLC doped with 790-nm CdSeTe QDs

633-nm excitation wavelength

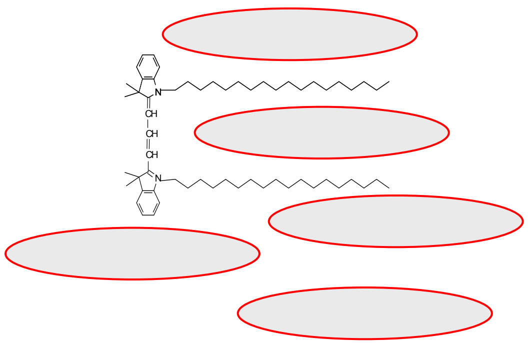


λ_{res_max} = 822 nm,
16 nm resonance bandwidth,
53-nm CdSeTe QD fluorescence bandwidth

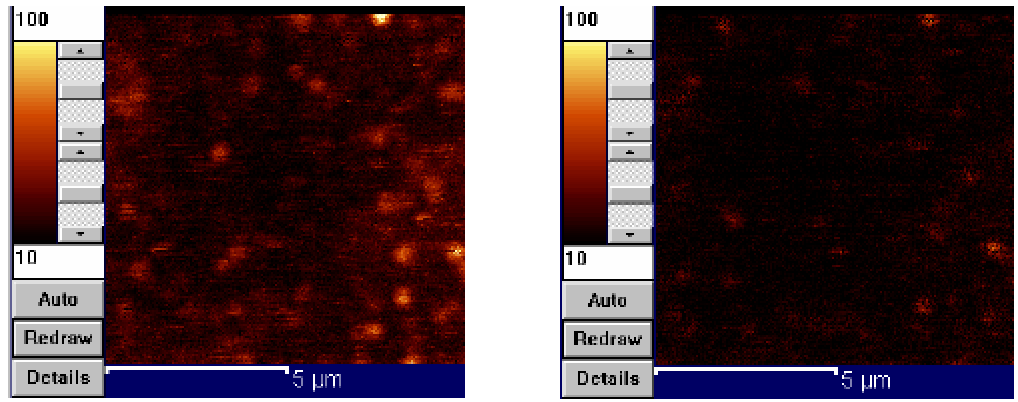
$g_e = 2(I_L - I_R)/(I_L + I_R)$

For unpolarized light $g_e = 0$; for circular polarized light $|g_e| = 2$.

Linearly polarized fluorescence of single DiI dye molecules in glassy nematic liquid crystal host



Perpendicular   Parallel



Figures clearly show that for this sample, the polarization direction of the fluorescence of single molecules is predominantly in the direction *perpendicular* to the alignment of liquid crystal molecules.

It is important that the background levels of left and right figures are the same.

S.G. Lukishova et al., *J. Mod. Opt.*, Special Issue on Single Photon: Sources, Detectors, Appl. and Measur. Methods, **54**, 417 (2007).

Summary

- **Several nonlinear optical effects under high-power nanosecond laser irradiation of liquid crystals were outlined:**

- **Nonlinear selective reflection:**
  athermal helical pitch dilation and unwinding of cholesteric mirrors by the field of a light wave

- **Nonlinear absorption and refraction:**
  (1) dependence of nonlinear refraction of liquid crystal on the laser beam diameter in presence of two-photon absorption,
  (2) cumulative effects in nonlinear absorption and refraction at low repetition rate (5-10 Hz) laser irradiation.

- **Feedback-free optical pattern formation**
  feedback-free kaleidoscope of patterns in dye-doped liquid crystals (hexagons/stripes etc.).

- **Cholesteric liquid crystal lasers and single-photon sources development was discussed.**

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Acknowledgments

Funding by the U.S. Army Research Office and National Science Foundation. Funding for graduate students by the Air Force and NASA

Graduate students

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University of Rochester Collaborators

Dr. A. Schmid, Prof. R. Boyd, Prof. C. Stroud,, Dr. K. Dolgaleva, Prof. S.-H. Chen, Dr. K. Marshall, Dr. S. Wei

Liquid Crystal Institute, Kent, OH

Thanks to Prof. P. Palffy-Muhoray for his hospitality and support, Prof. T. Kosa and Prof. B. Taheri for their help.

Moscow Institute of Radioengineering and Electronics, Russ.Acad Sci.

Collaboration with Prof. S. Belyaev and Dr. N. Malimonenko (Moscow Organic Intermediates and Dyes Institute), students of Moscow Institute of Physics and Technology K. Lebedev and E. Magulariya.