Interplay of defects in liquid crystals & optical vortices in beams of light

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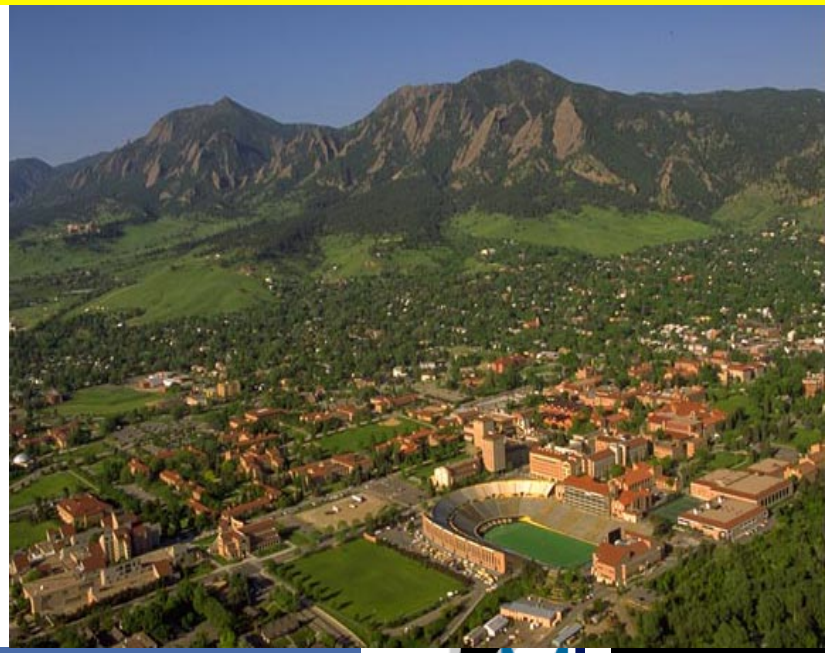

Colorado Physics
Department of Physics•University of Colorado at Boulder

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MRLC


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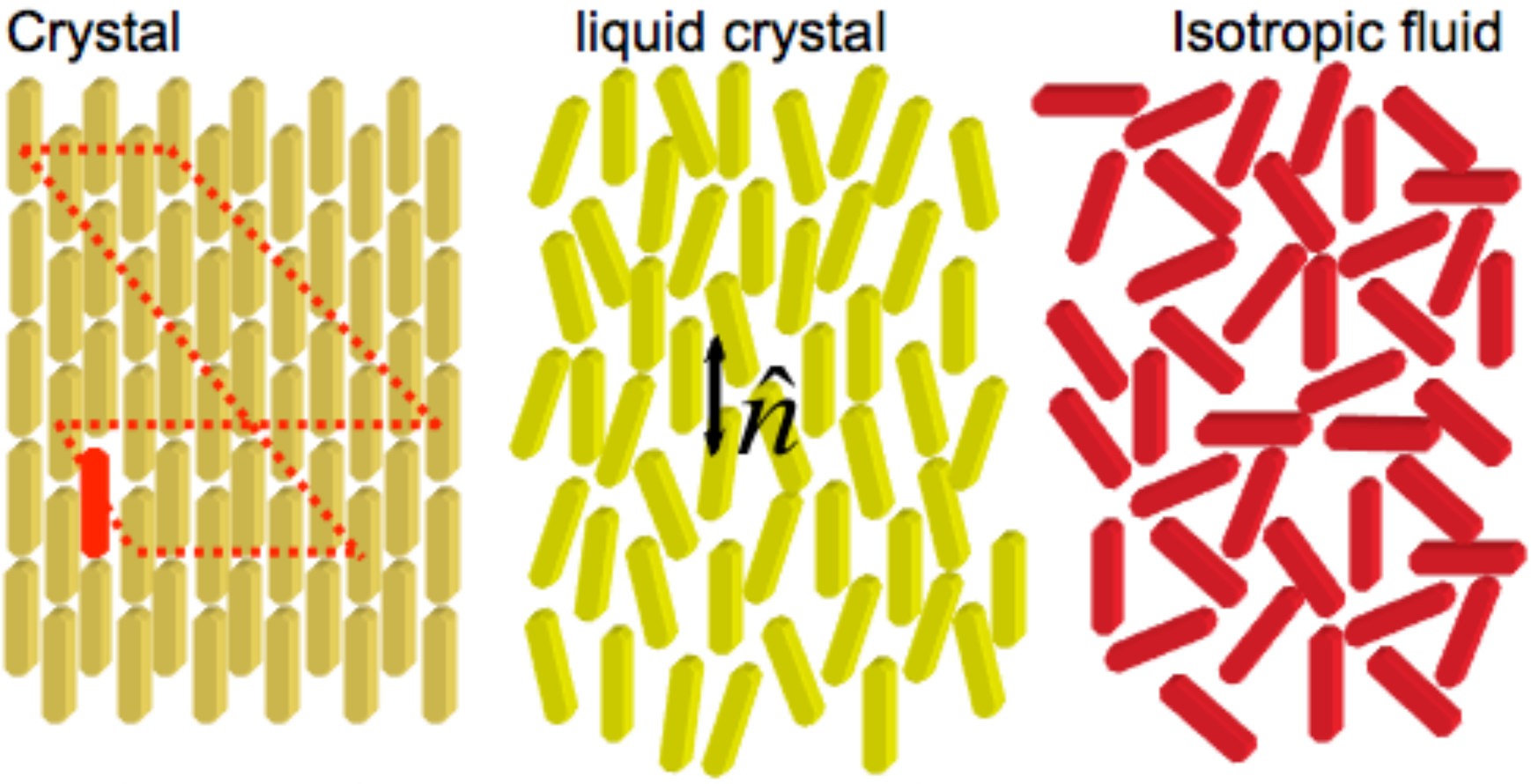
Outline

- Simultaneous 3D imaging & manipulation of director field **n**(**r**);
- Optical generation of twist-bound configurations of long-term stable defects;
- Torons, Skyrmions, Merons *et al* in LCs;
- Crystals & quasicrystals formed by defects;
- Control of matter defects by optical vortices;
- Control of optical vortices by dislocations in crystals of multi-scale defects;
- Self-assembly and layer-by layer optical patterning of defect configurations.

Nematic Liquid Crystals (LCs)

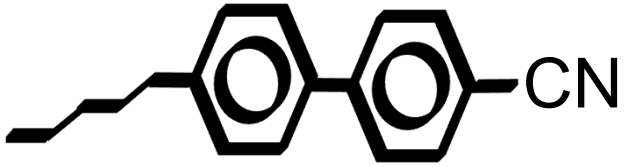
→Flow like liquids;
→Anisotropic like solid crystals;



Average local molecular orientations in liquid crystals are described by the director with head-tail symmetry $\hat{n} \equiv -\hat{n}$

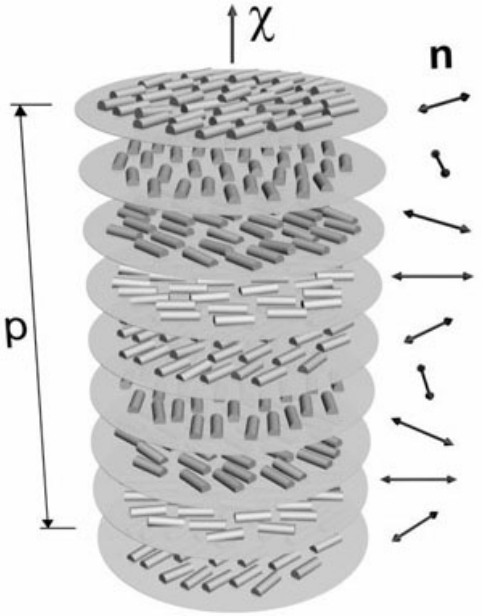
Chiral LCs: twisted ground states

Chiral & ordinary nematic LCs:

5CB 

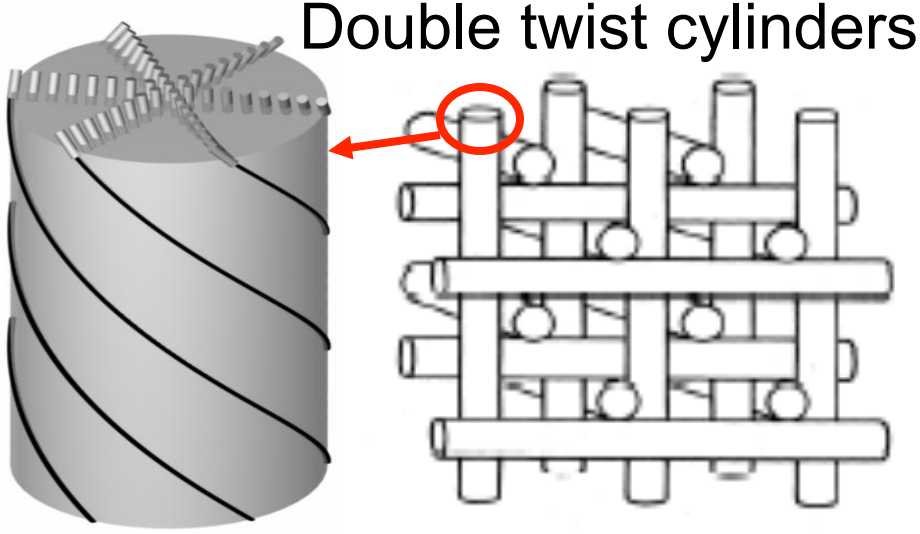
CB15 

Cholesteric phases

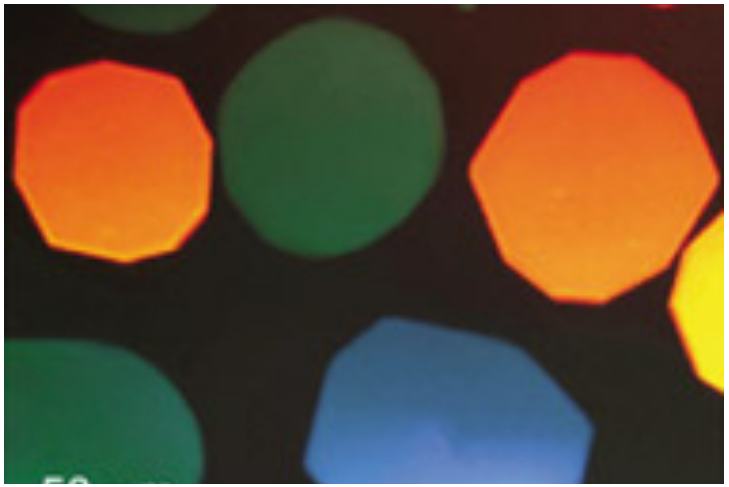


Blue phases

Double twist cylinders



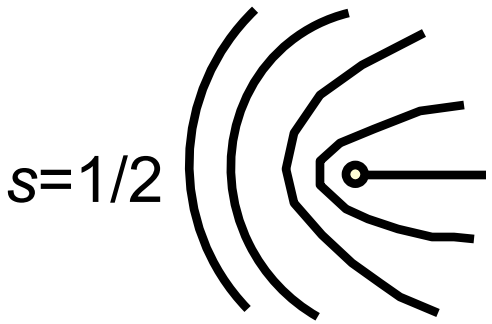
Periodicity can be ~100nm

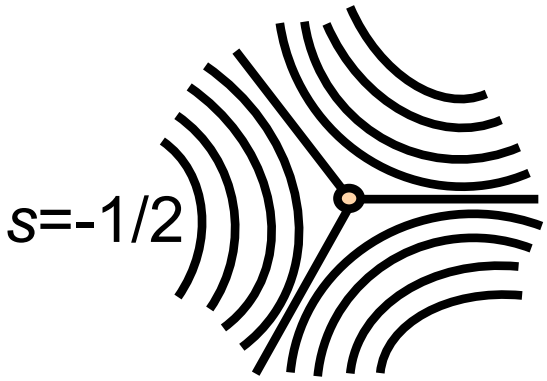


Coles et al., *Nature* **436,** 997 (2005)

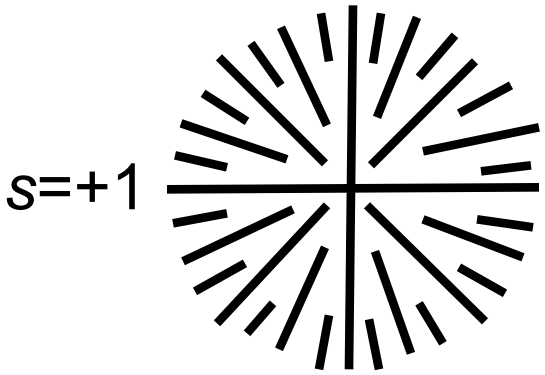
W. Cao et al., Nature Materials, 111, (2002).

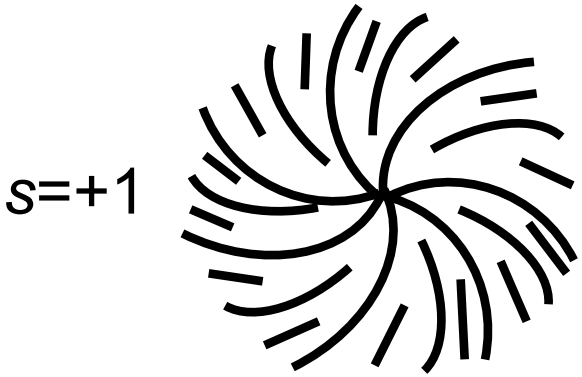
Defects in Nematic LCs: Disclinations

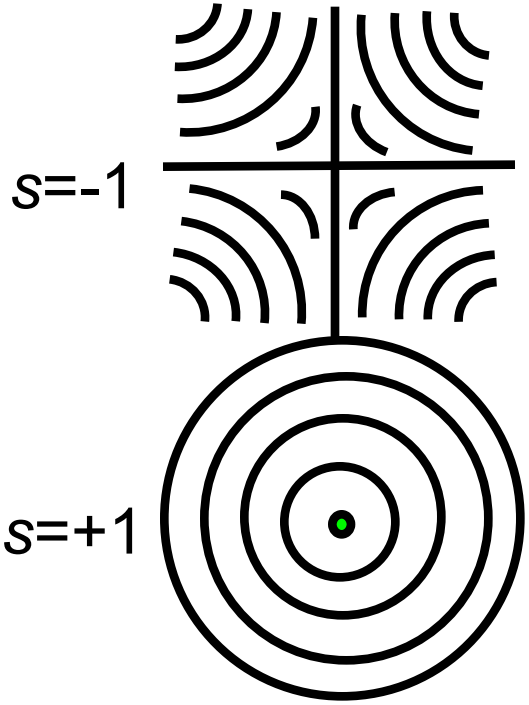


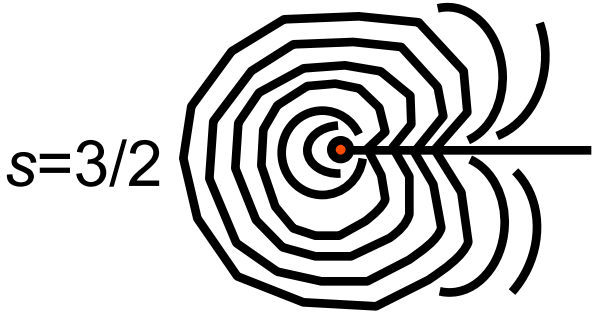


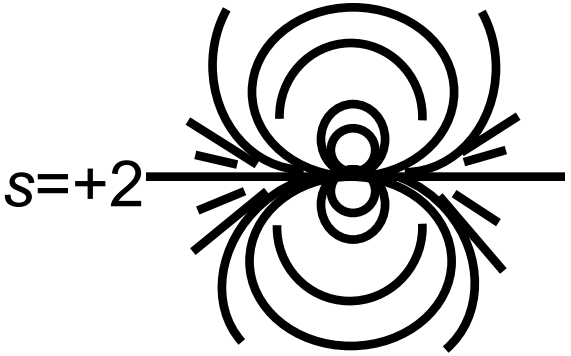
The singular line (disclination) is pointing out of the page, & director orientation changes by $2\pi s$ on going around the line (s is the strength)



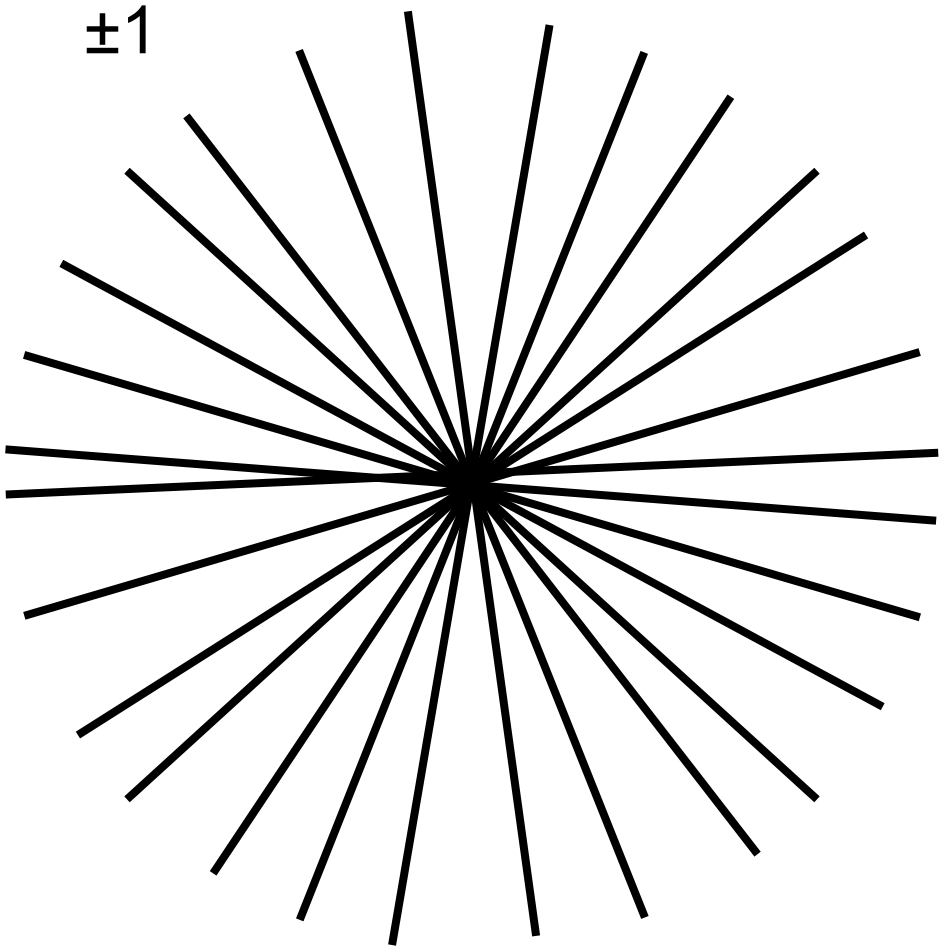




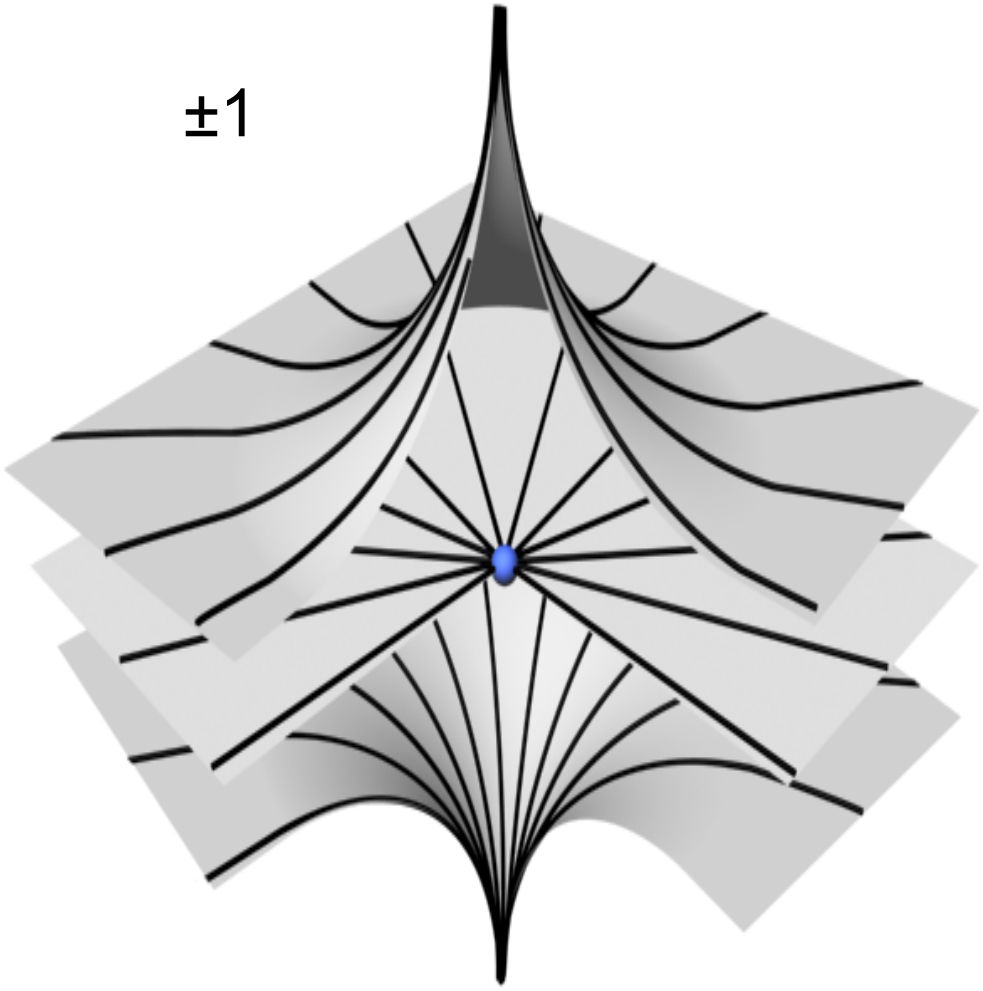




Point Defects



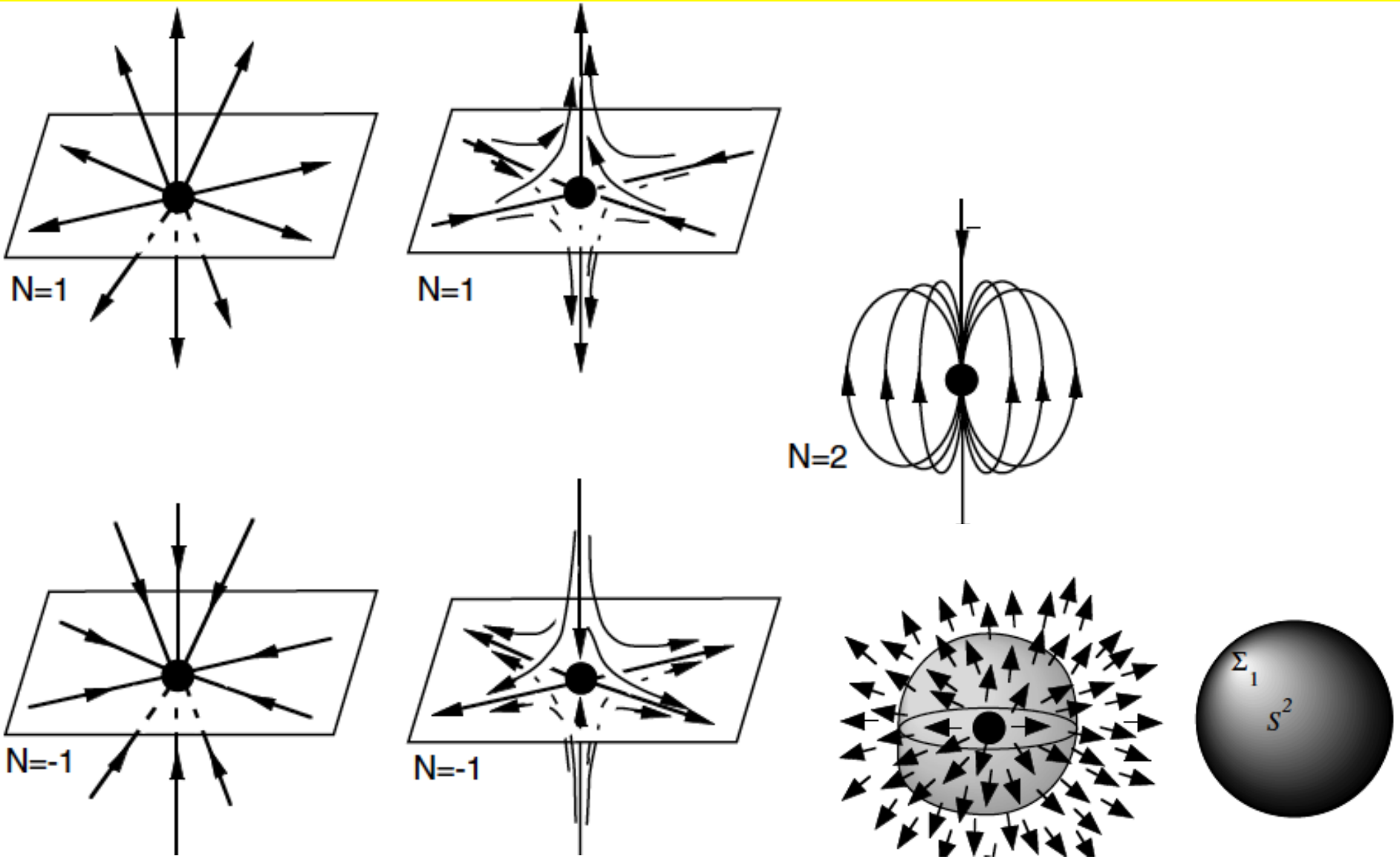
- Radial Hedgehog



- Hyperbolic

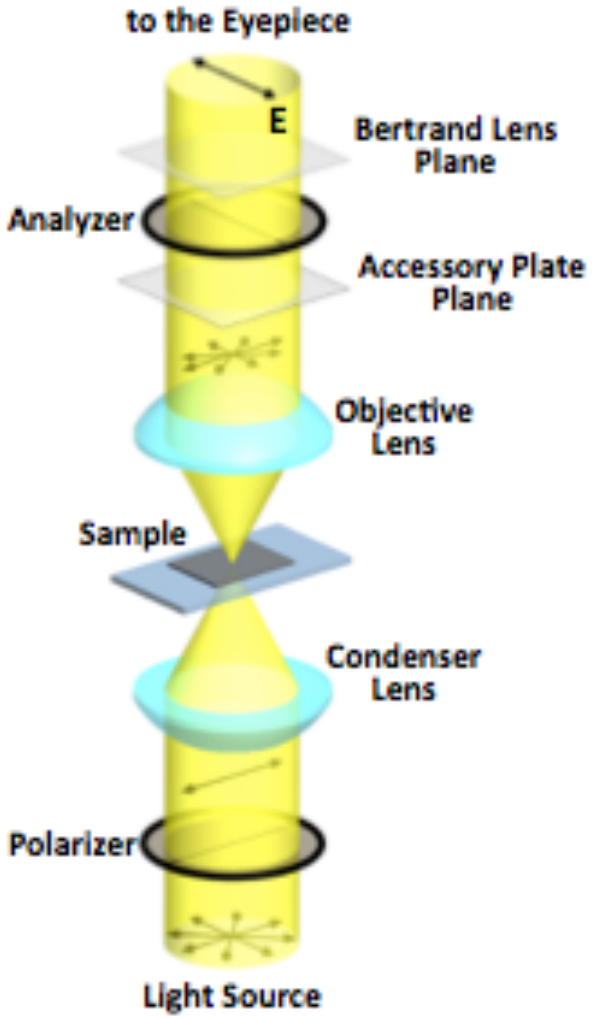
Alexander, Chen, Matsumoto, Kamien, "Disclination Loops, Hedgehogs, and All That" Rev. Mod. Phys. **84** (2012) 497

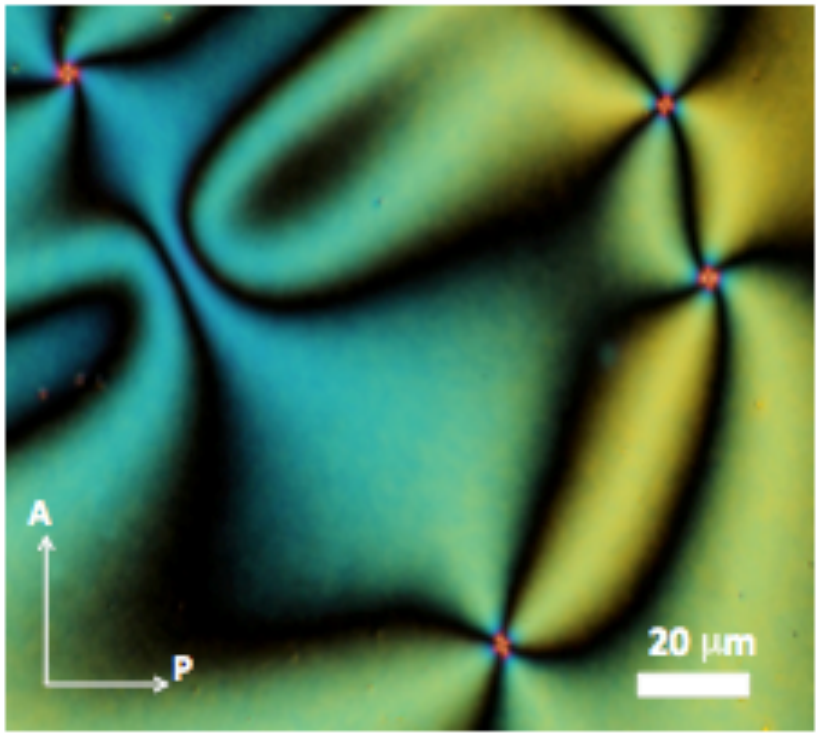
Point defects in a 3D vector field



How many times they cover the S^2 order parameter space?

Polarizing optical microscopy

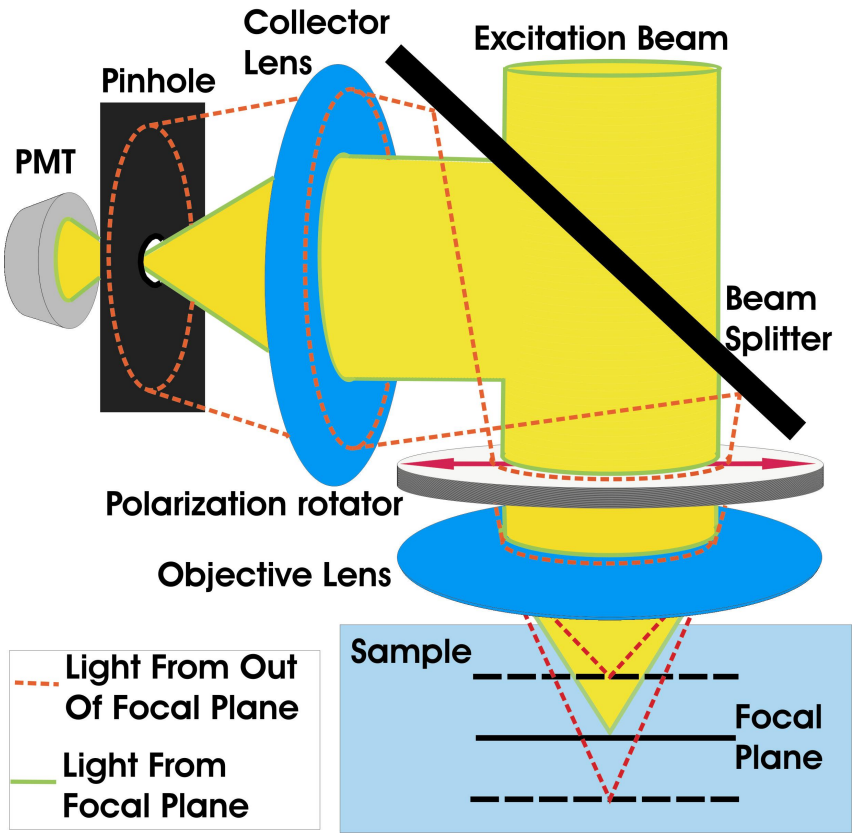




$$I_{PM}(x,y) = I_0 \sin^2 2\beta \sin^2\left(\frac{\pi}{\lambda}\int_0^h \Delta n_{eff}(x,y,z)dz\right)$$

- Director is a coordinate-dependent optical axis;
- 3D field from 2D picture?..
- Works well only with 2D z-independent textures!
- Once the configuration becomes nontrivial, the technique stops working

Fluorescence Confocal Polarizing Microscopy

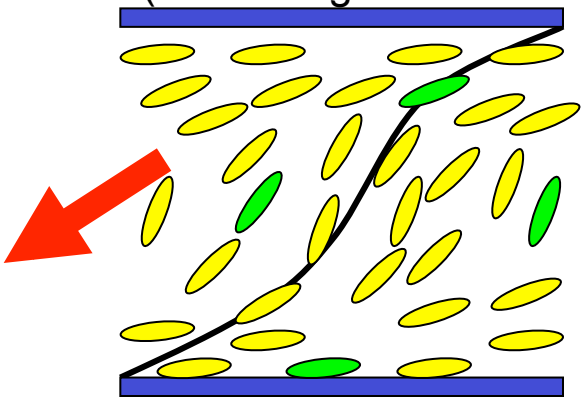


Basic features:

1) polarized excitation and fluorescence detection

2) fluorescence dye alignment

(as in the guest-host LCD)

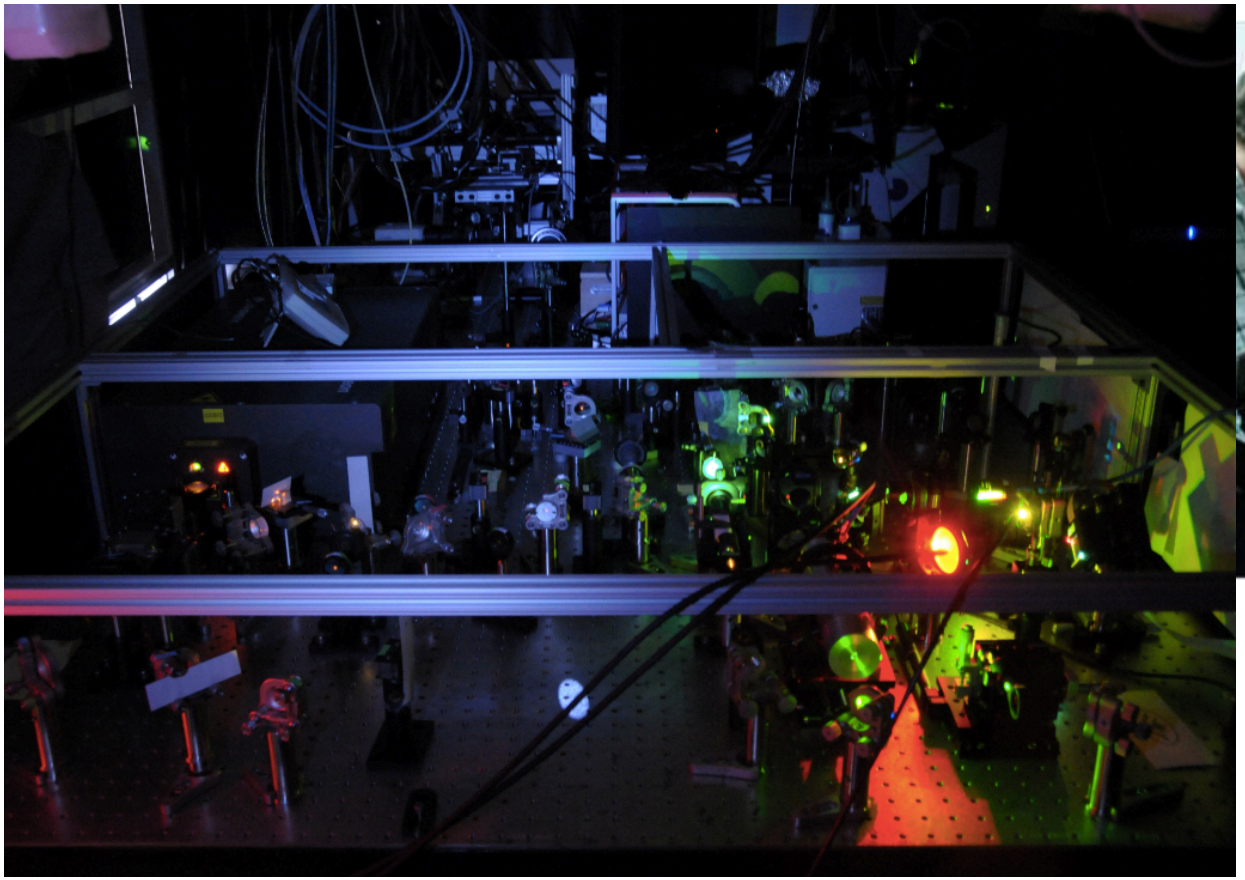


- Requires doping with special dyes (not always possible)
- Limitations in imaging Biaxial director fields…

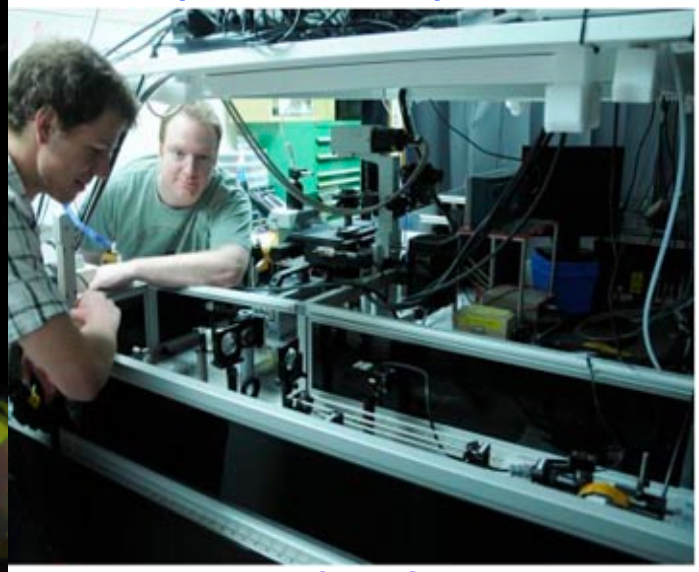
I.I. Smalyukh, S.V. Shiyanovskii, and O.D. Lavrentovich, *Chem. Phys. Lett*. **336**, 88 (2001).

Multimodal nonlinear optical imaging of LCs

→3D linear/nonlinear optical imaging



→3D optical manipulation



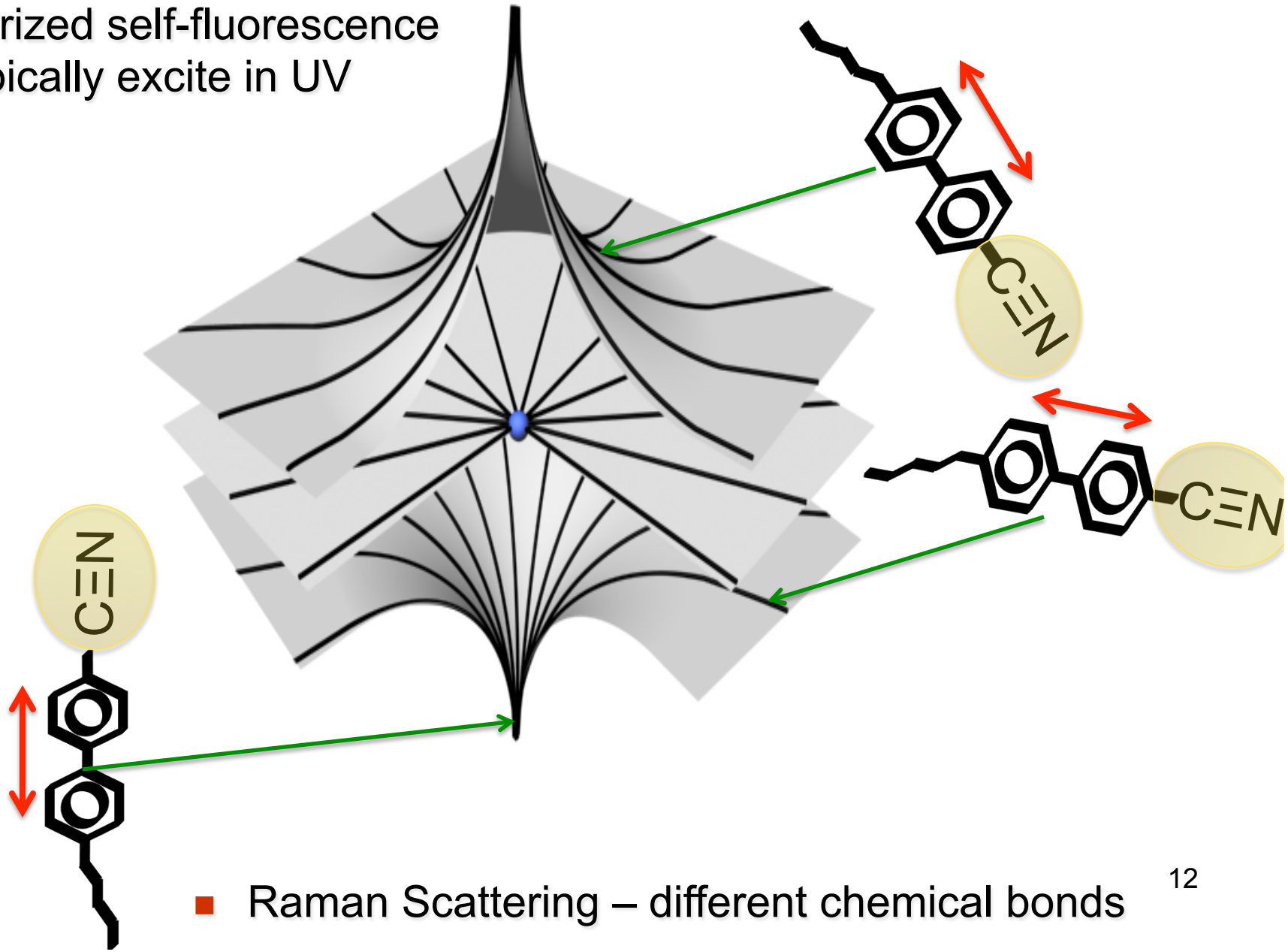
→Broadband CARS-PM
→Broadban SRS-PM
→Multiphoton fluorescence
→Multiple harmonic generation
→Holographic Optical Tweezers

P.G. de Gennes ”...the study of liquid crystals is complicated because it involves a certain sense of vision in three-dimensional space in order to visualize complex molecular arrangements…” in the book ”The Physics of Liquid Crystals”

→Label-free chemically-specific orientationally-sensitive 3D imaging
→Simultaneous with non-contact optical manipulation in 3D
→Many imaging modalities with complementary capabilities

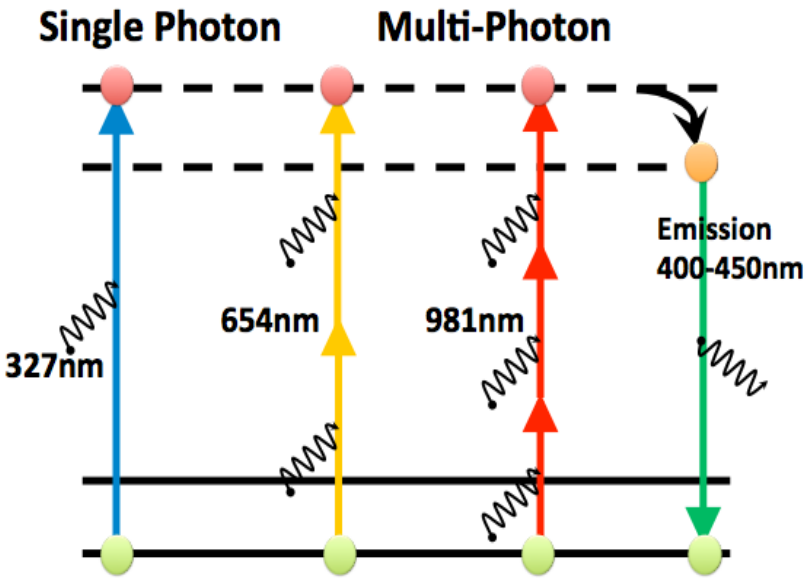
Label-free study of the director fields in 3D?

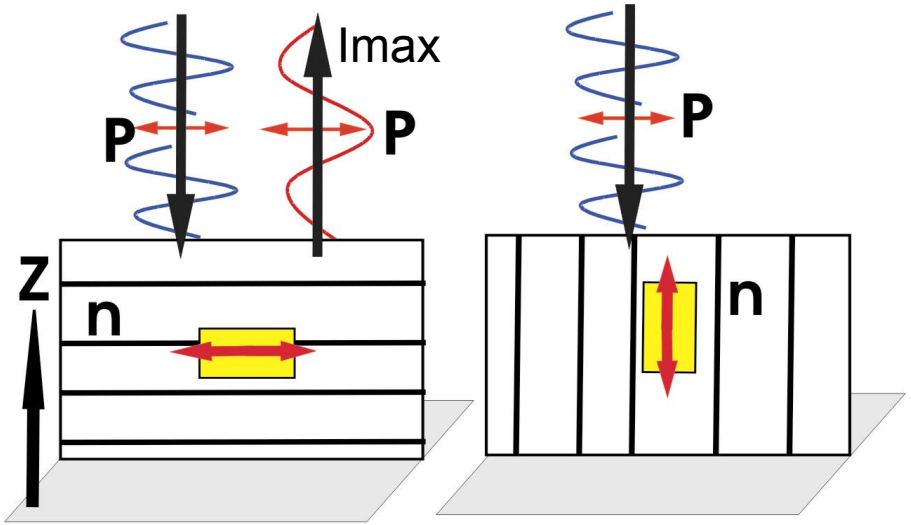
- Polarized self-fluorescence
  – typically excite in UV

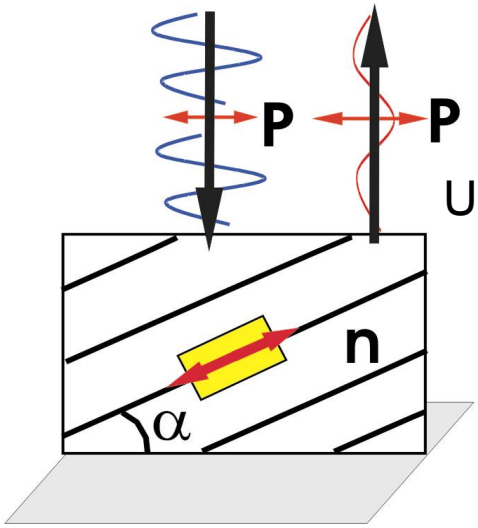


- Raman Scattering – different chemical bonds

Multiphoton self-fluorescence imaging: no dyes







Unpolarized detection:

$$I_{em} \propto I_0 \cos^{2i} \alpha$$

Polarized detection:

$$I_{em} \propto I_0 \cos^{2(i+1)} \alpha$$

- Strong orientational sensitivity!

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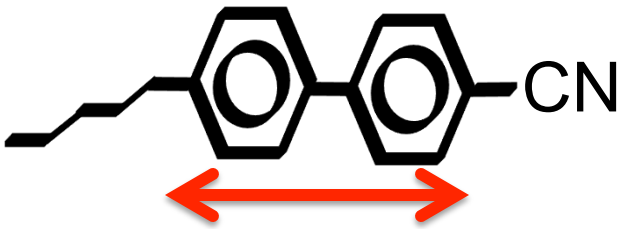
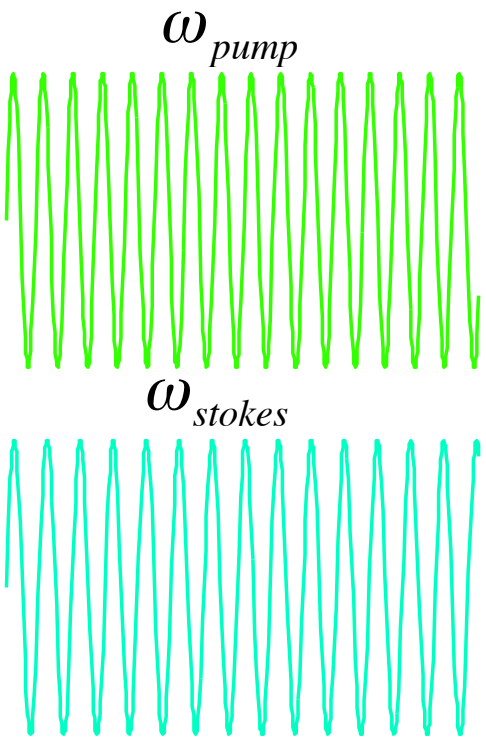


Absorption/emission transition dipole

T. Lee, R.P. Trivedi, & I.I. Smalyukh, *Opt. Lett.* 35, 3447-3449 (2010).

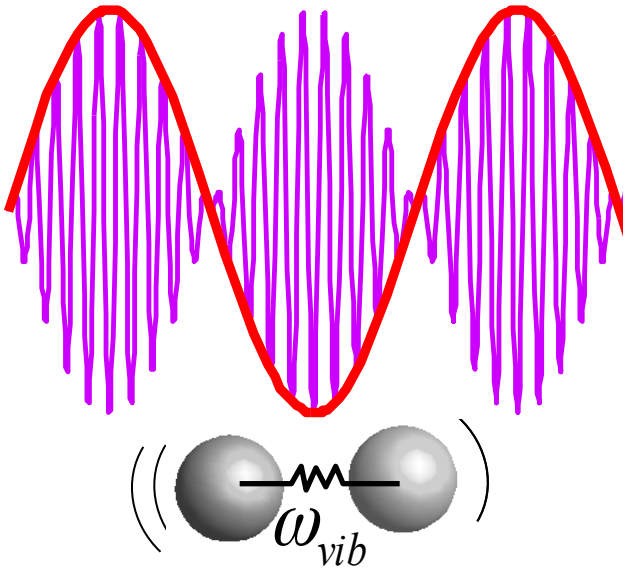
Coherent anti-Stokes Raman Scattering (CARS)

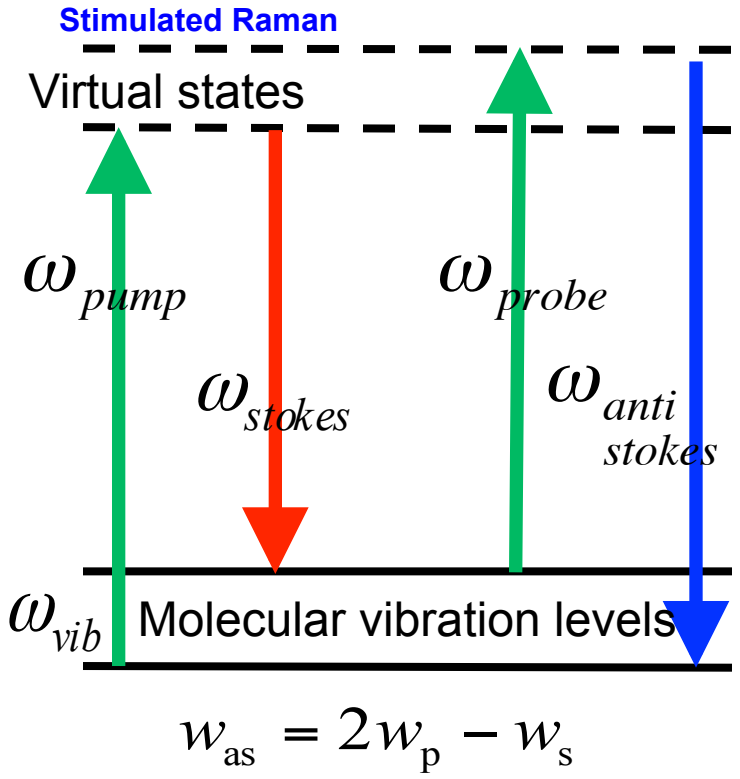
→ CARS uses two laser frequencies to interact resonantly with a specific molecular vibration;



Beating at:

$$\omega_{pump} - \omega_{stokes} = \omega_{vib}$$





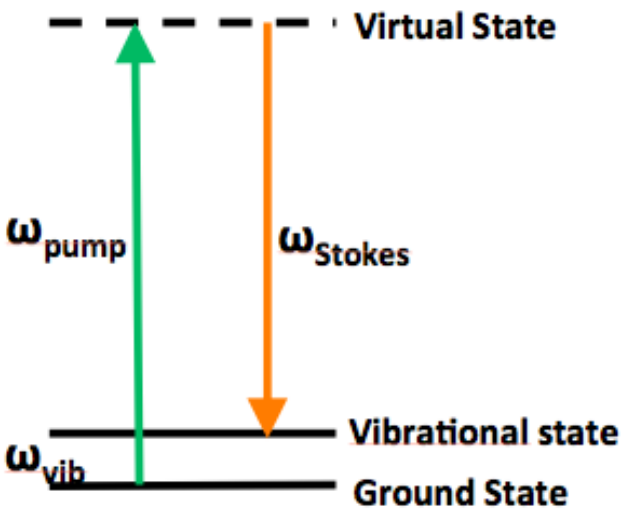
$$w_{as} = 2w_p - w_s$$

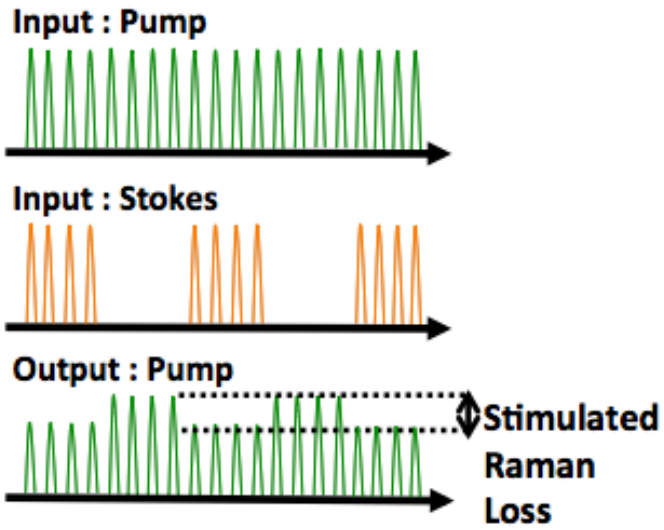
→ 3rd order nonlinear process with independent pump/probe and Stokes lasers

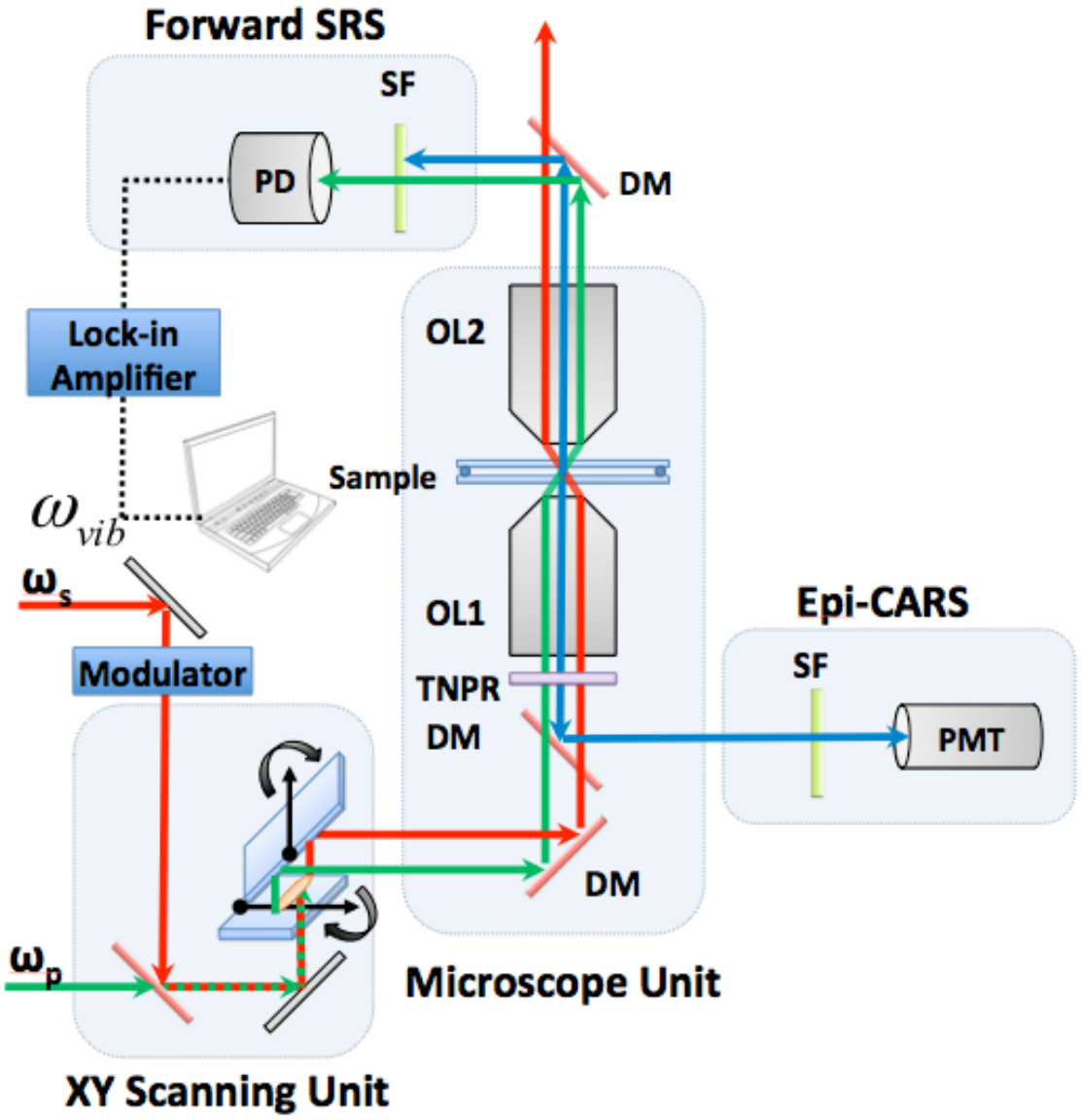
→ When the beat frequency matches the frequency of a particular Raman vibration, a strong anti-Stokes signal is generated at signal at $\omega_{as}=2\ \omega_p - \omega_S$

→ Broadband Stokes excitation & CARS detection with Spectral shaping;

Stimulated Raman scattering polarizing m-py



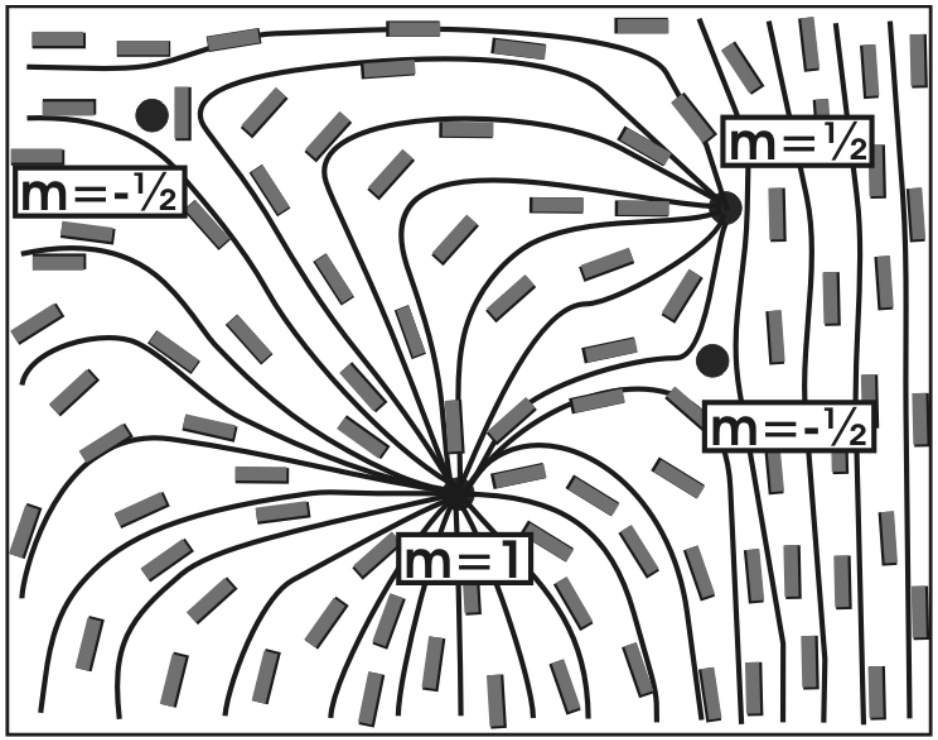


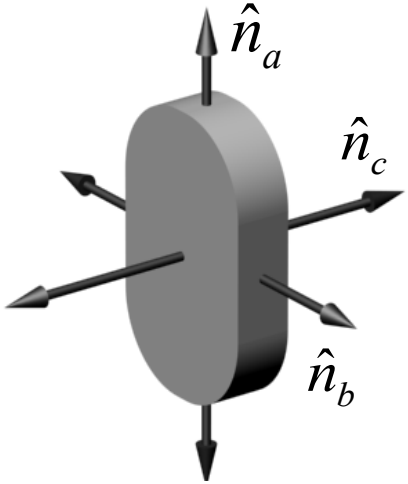


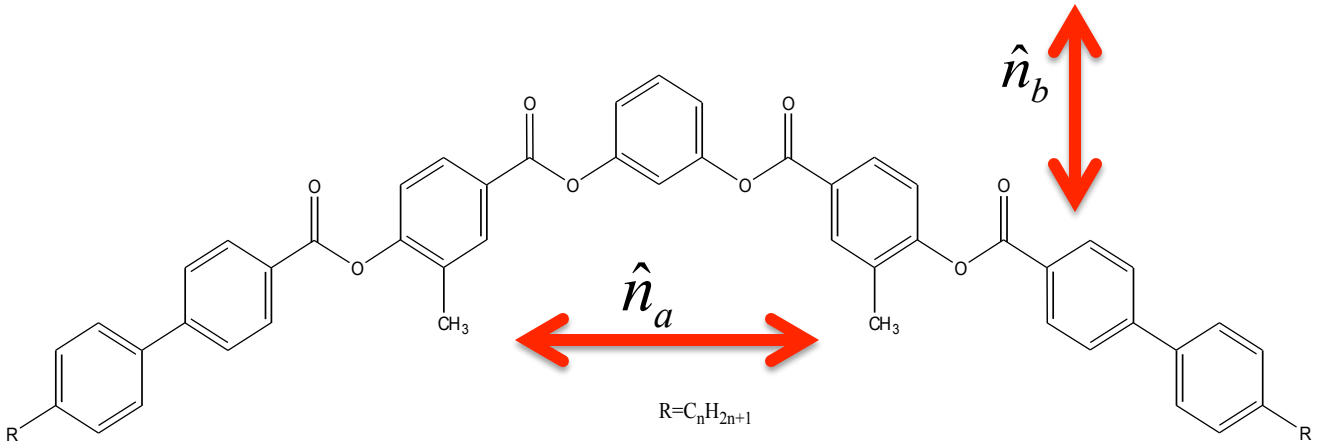
→ 2nd order nonlinear process with independent pump/probe and Stokes lasers

→ Modulate the Stokes beam – measure the Stimulated Raman Loss signal

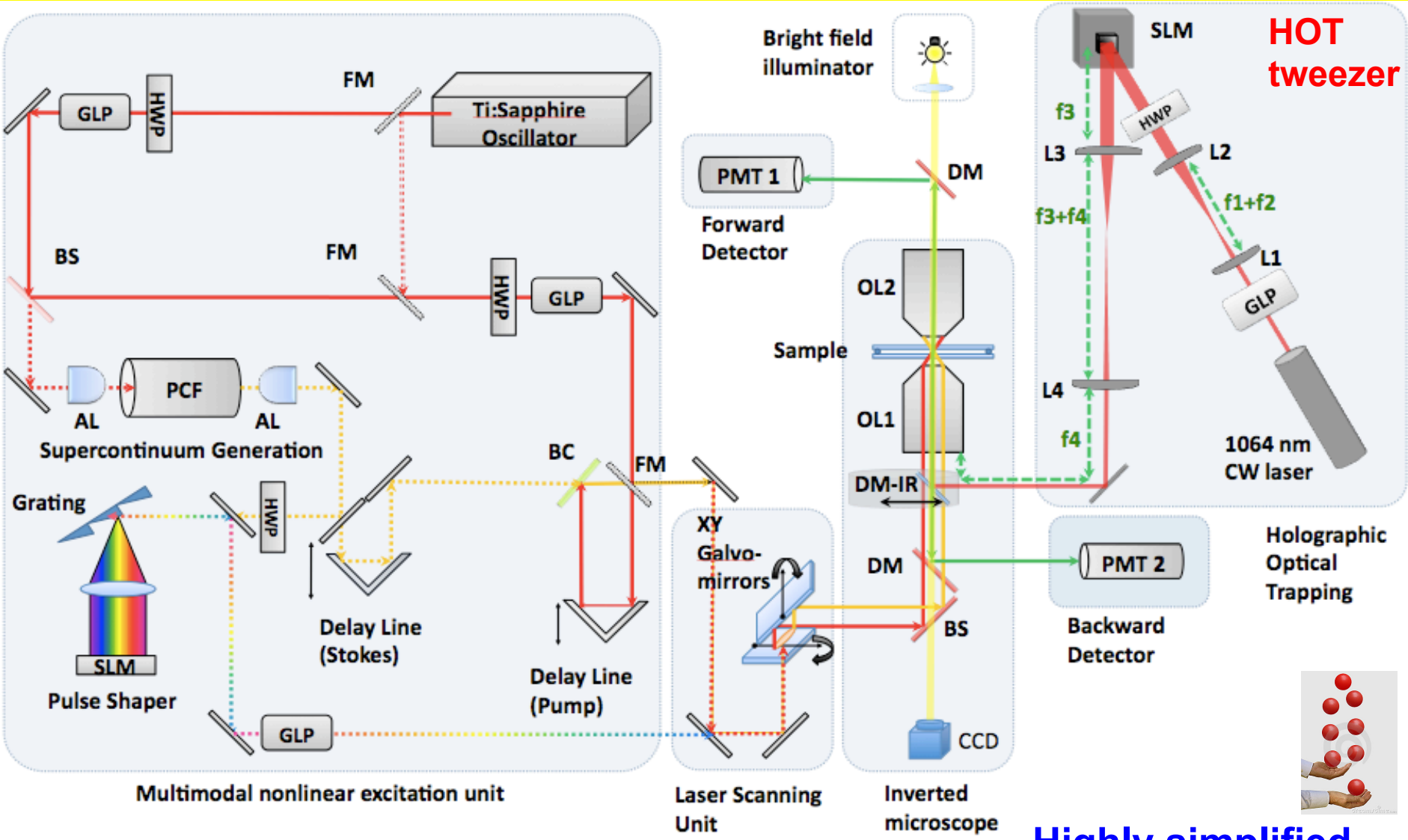
CARS-PM imaging of director field in biaxial LCs







Integrated holographic optical tweezers & multimodal 3D imaging: setup schematic



Highly simplified schematic

T. Lee, R. P. Trivedi & I. I. Smalyukh, *Opt. Lett.* 35, 3447 (2010);
R.P. Trivedi, T. Lee, K. Bertness, & I.I. Smalyukh, *Opt. Express* 18, 27658-27669 (2010).

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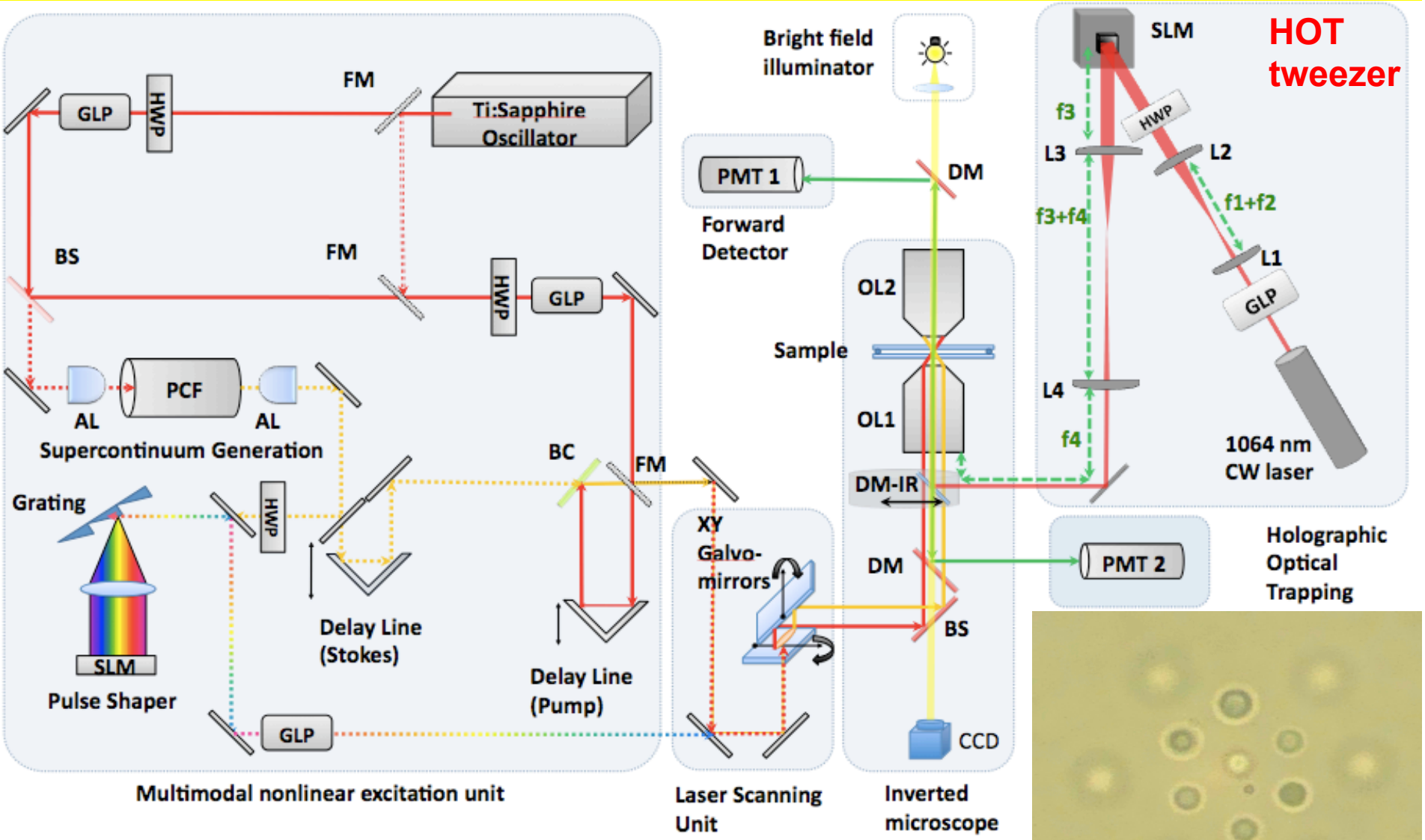

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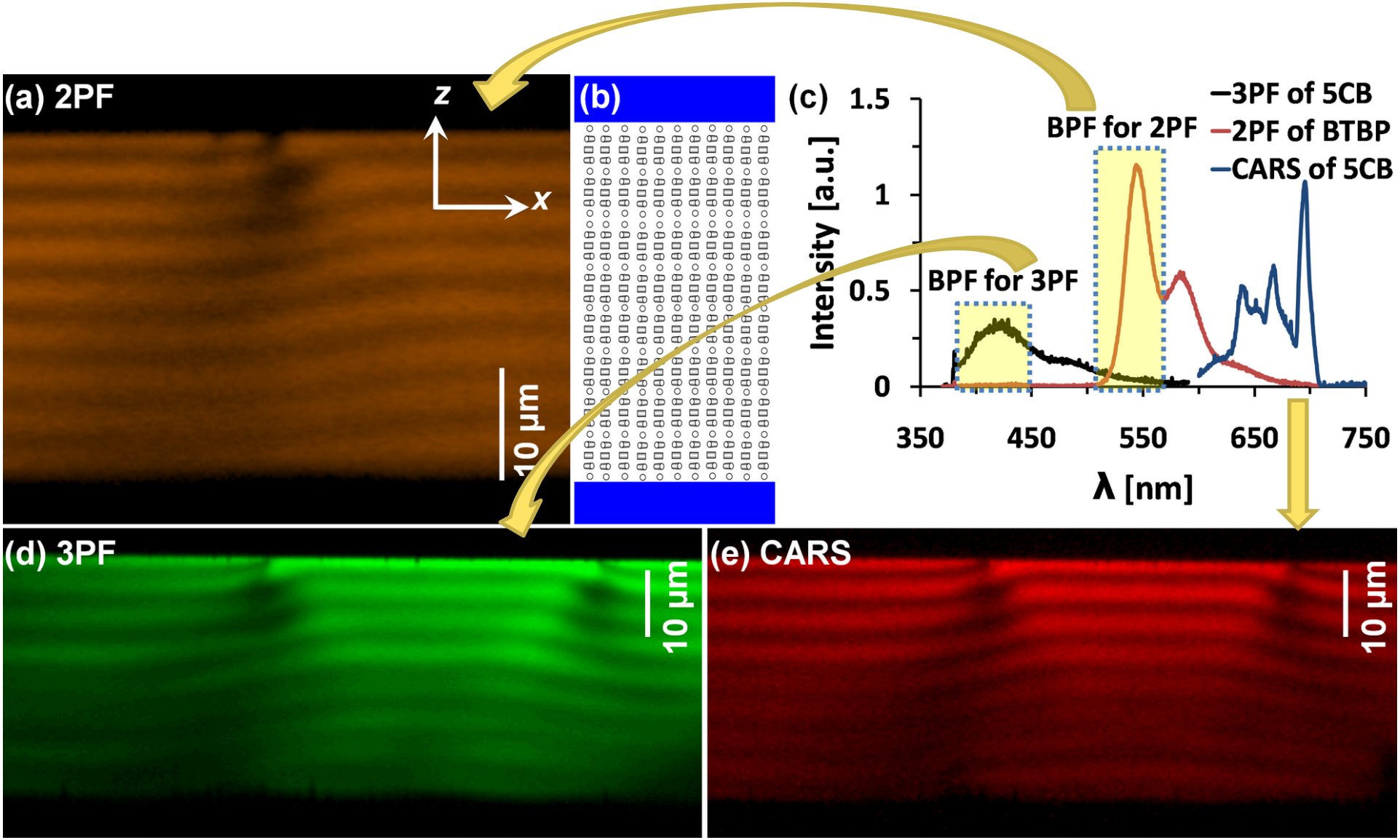
R.P. Trivedi, T. Lee, K. Bertness, & I.I. Smalyukh, *Opt. Express* 18, 27658-27669 (2010).

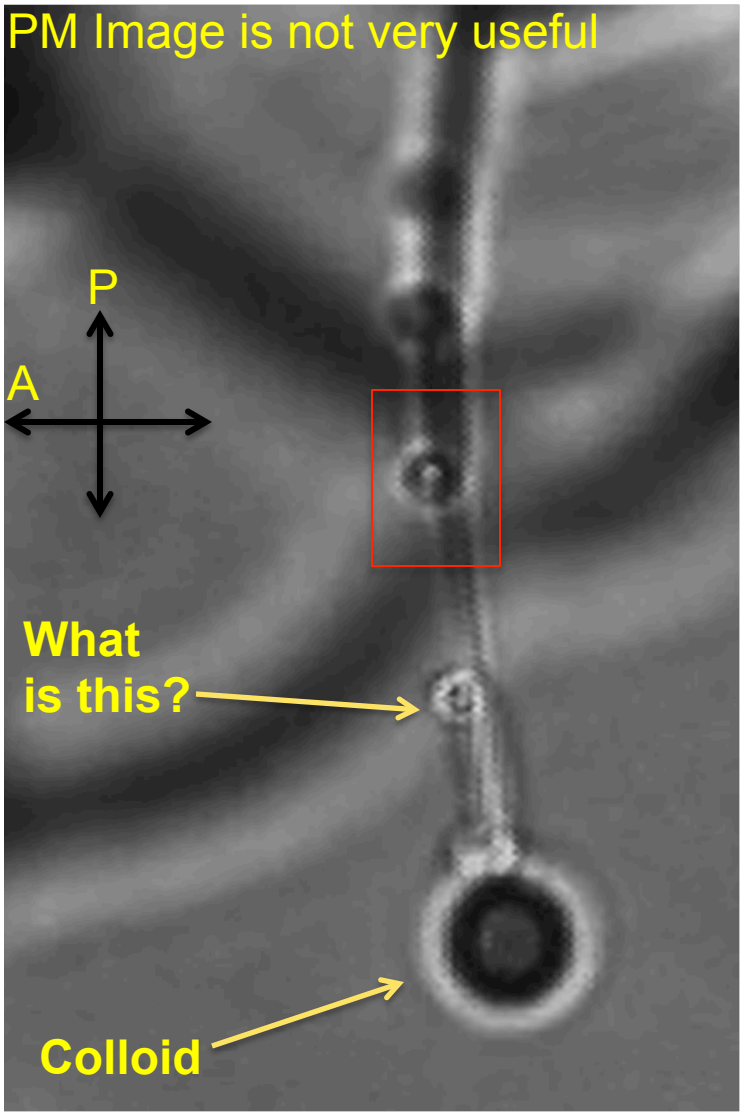
Simultaneous 3D imaging in different modalities



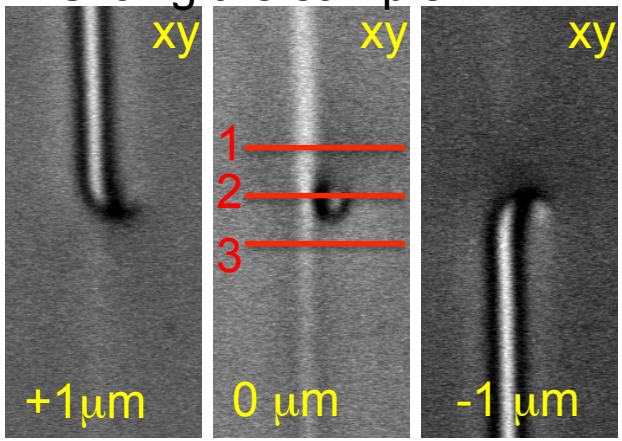
Trivedi, Lee, Bertness, & Smalyukh, *Opt. Express* **18**, 27658-27669 (2010).

Simultaneous 3D “drawing” & imaging of defects

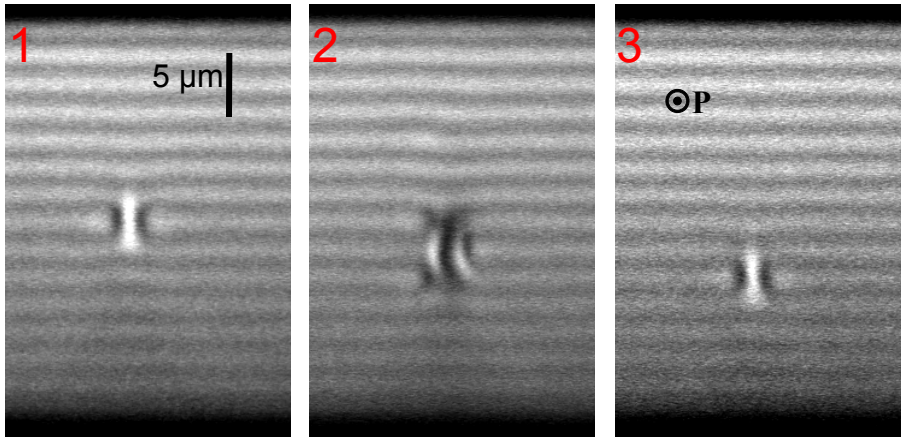
3D drawing of defects by a colloid



Slicing the sample



Slicing Vertically Across the defect



Trivedi, Senyuk, Lee, Smalyukh, PNAS **109**, 4744-4749 (2012).

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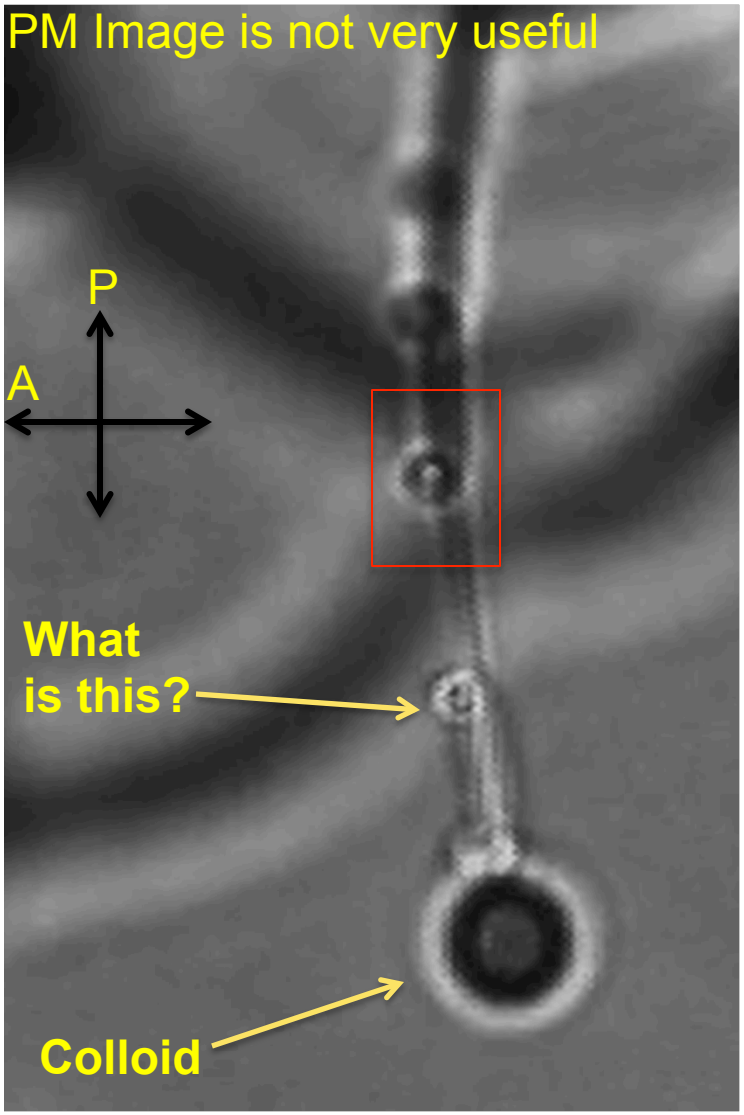

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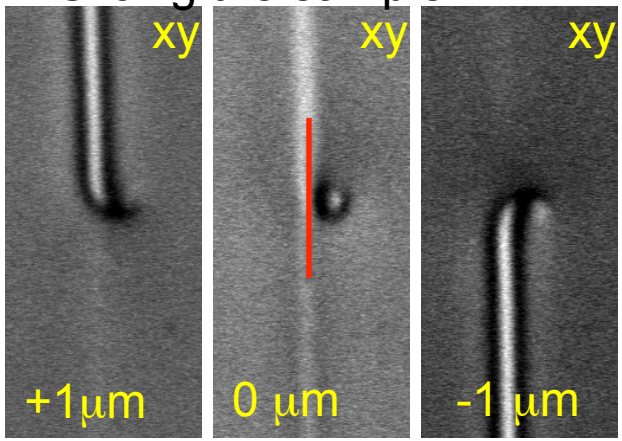
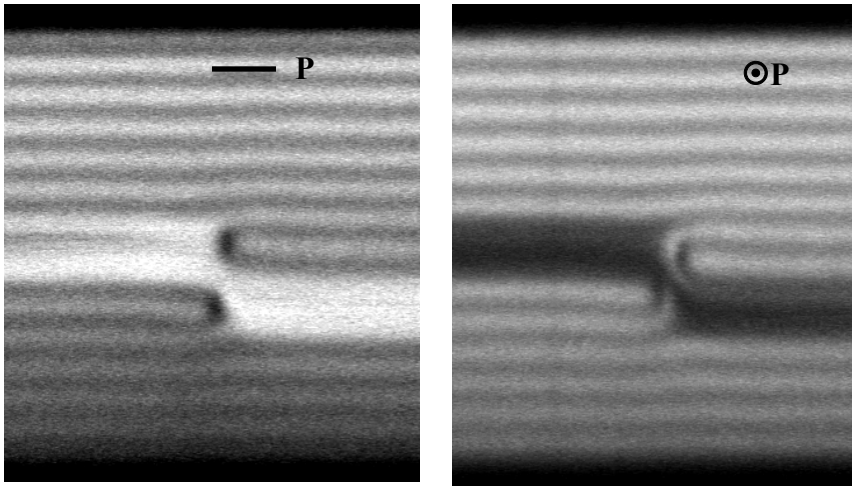


Slicing Vertically along the defect



Trivedi, Senyuk, Lee, Smalyukh, PNAS **109**, 4744-4749 (2012).

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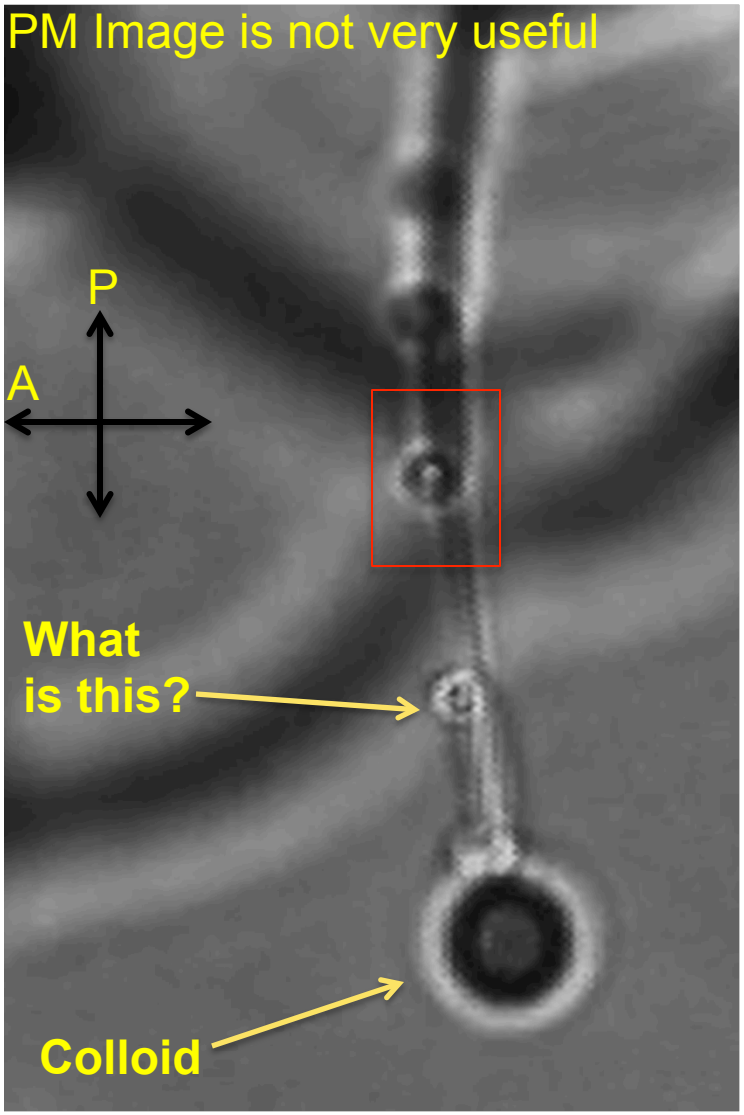

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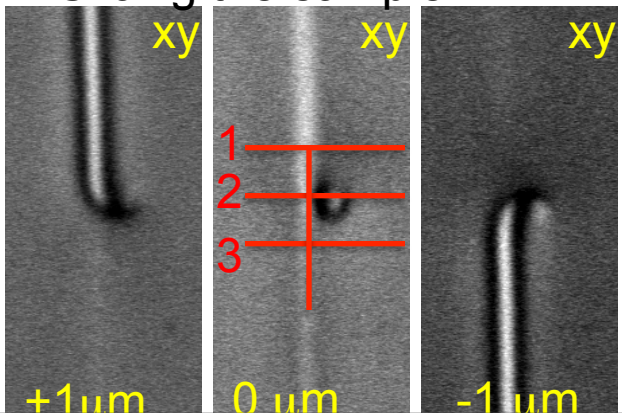
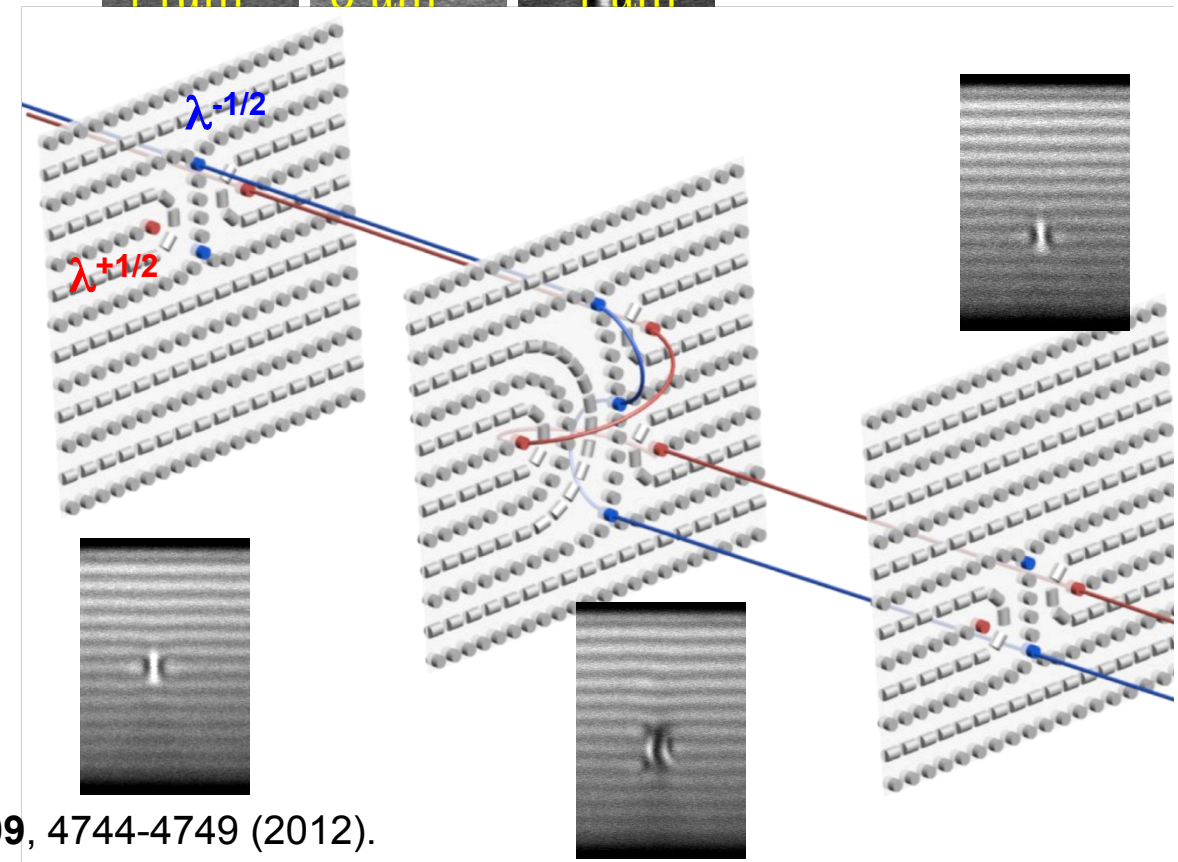
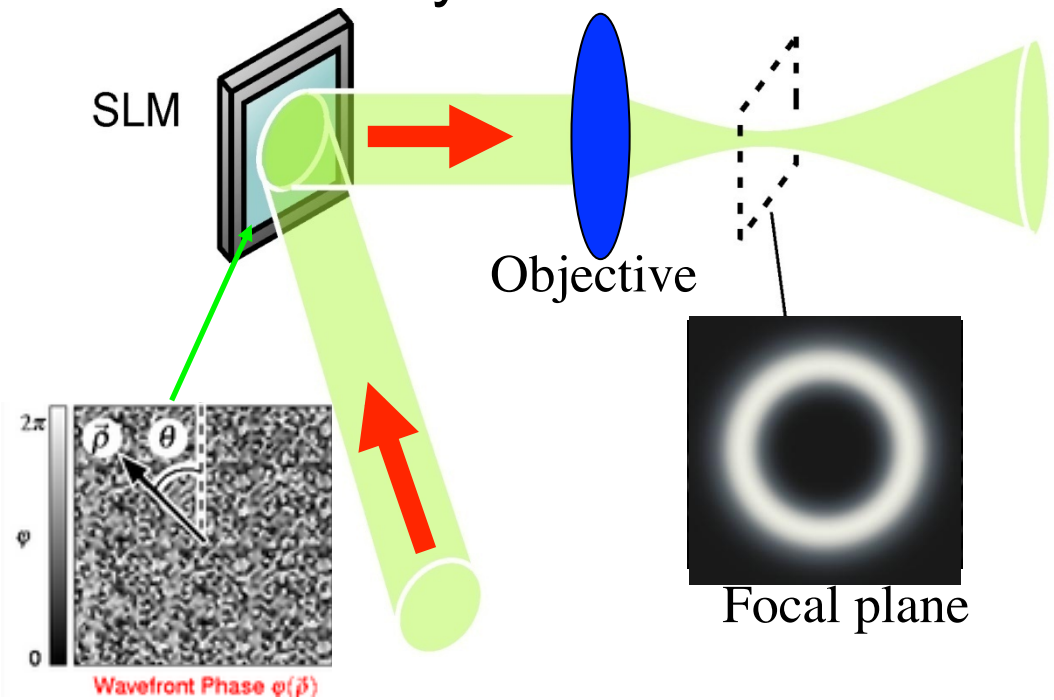




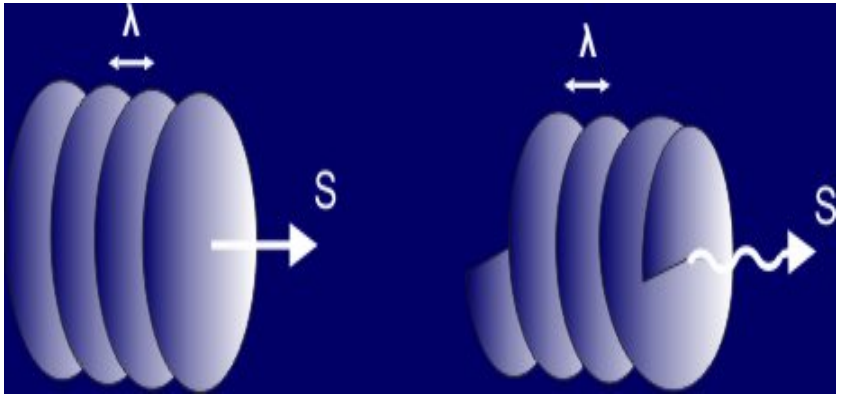
Trivedi, Senyuk, Lee, Smalyukh, PNAS **109**, 4744-4749 (2012).

Beam shaping using a spatial light modulator

Phase-only LC SLM

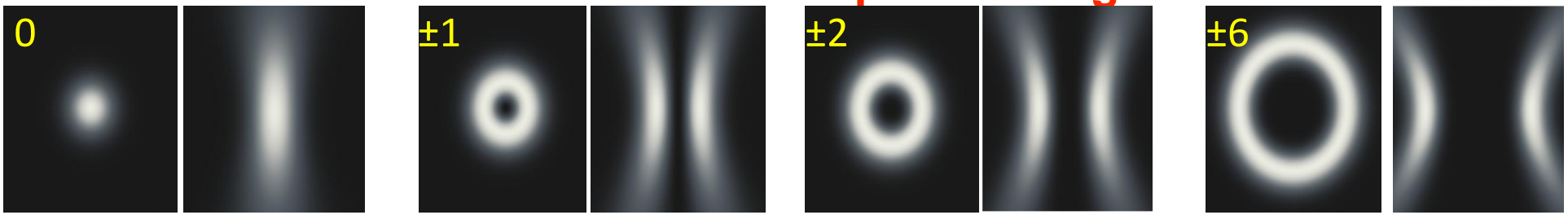


Optical vortices - phase singularities - screw dislocations



*L. Allen et al., PRA **45**, 8185 (1992)*

•Screw dislocations in the phase of light



J. Courtial et al., Phys Rev Lett 80, 3217, 1998

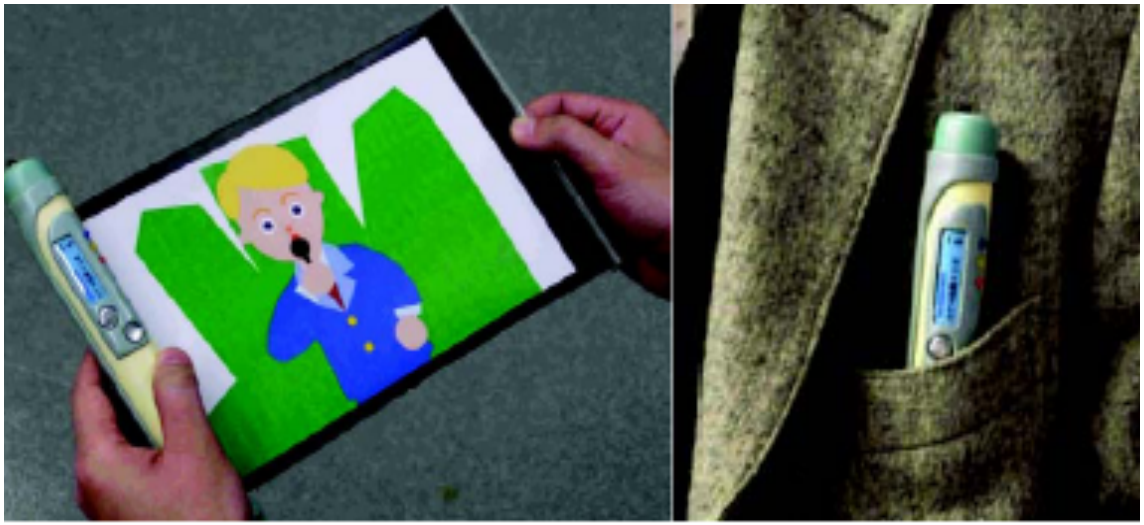
Can optical vortices control LC defects/fields?

Electrically-Controlled LC director

→Facile collective response to external fields;

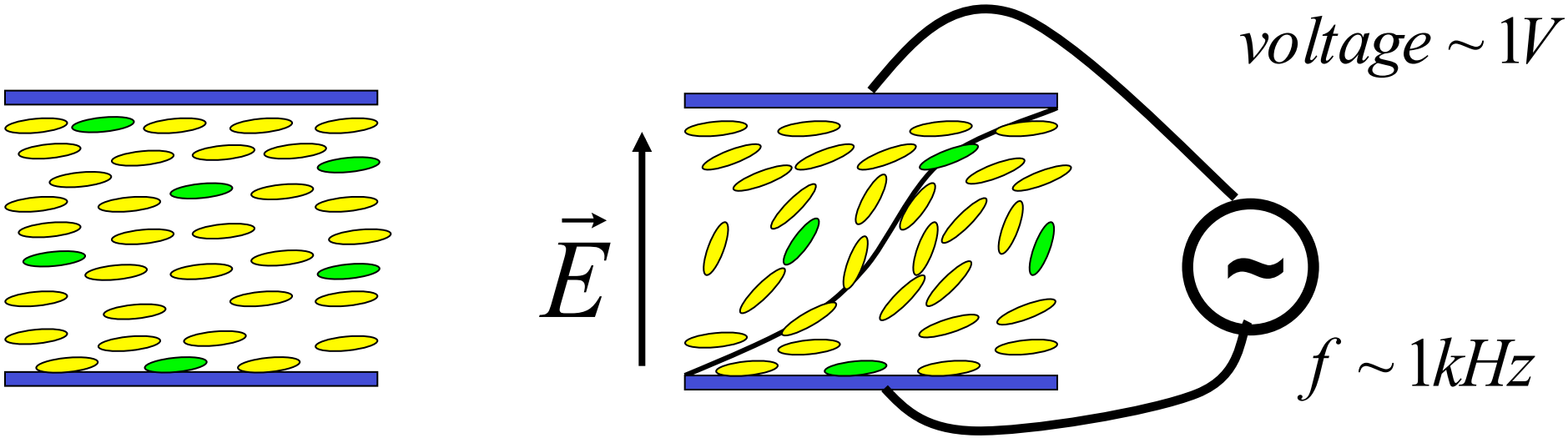
→Electric-field-induced realignment in LCs;

→Minimize the electric-field term of free energy:



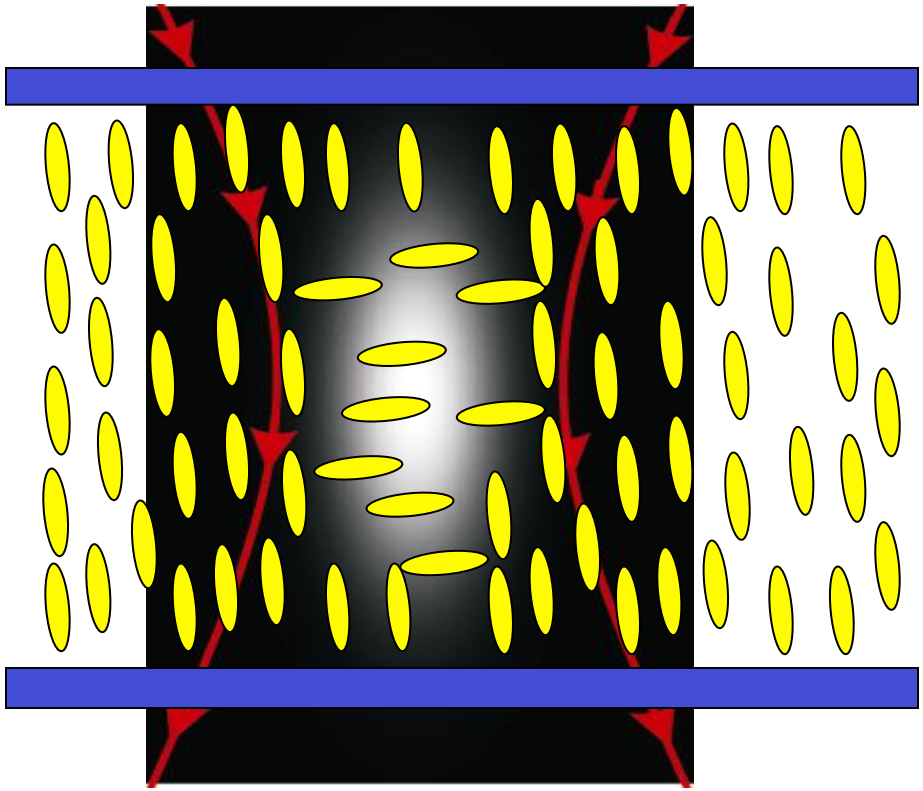
→Displays and electro-optic devices

$$F_e = -\frac{\varepsilon_0}{2}\int_V \Delta\varepsilon(\vec{E}\cdot\hat{n})^2 dV$$

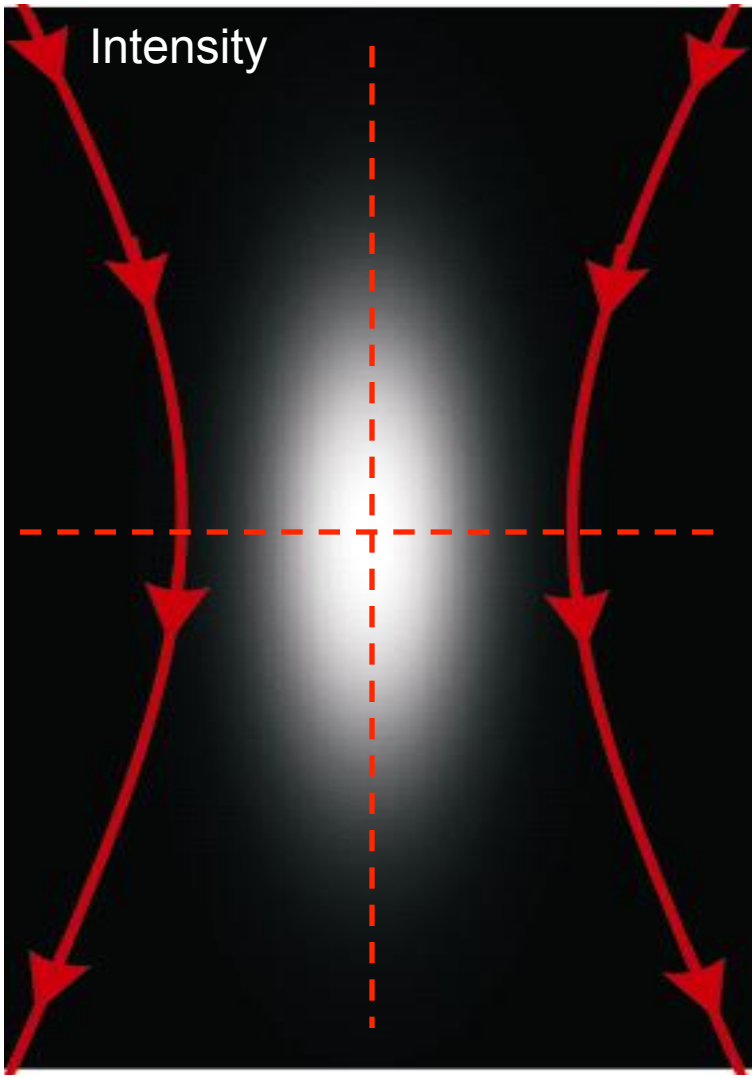


Optically-induced LC director realignment

Optical realignment



Focused Gaussian beam



$Intensity \sim E^2$

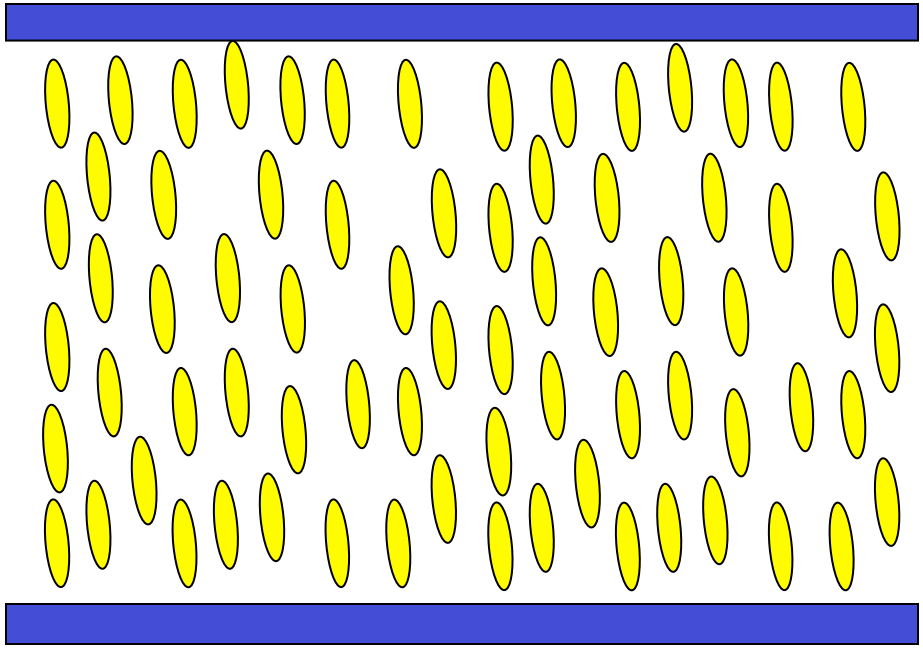
→Realignment by electric field of laser beam;

$$F_e = -\frac{\varepsilon_0}{2}\int_V \Delta\varepsilon(\vec{E}\cdot\hat{n})^2 dV$$

→At optical frequencies $\Delta\varepsilon = n_e^2 - n_o^2$

Optically-induced director realignment

Once Laser is turned off – realignment to uniform state



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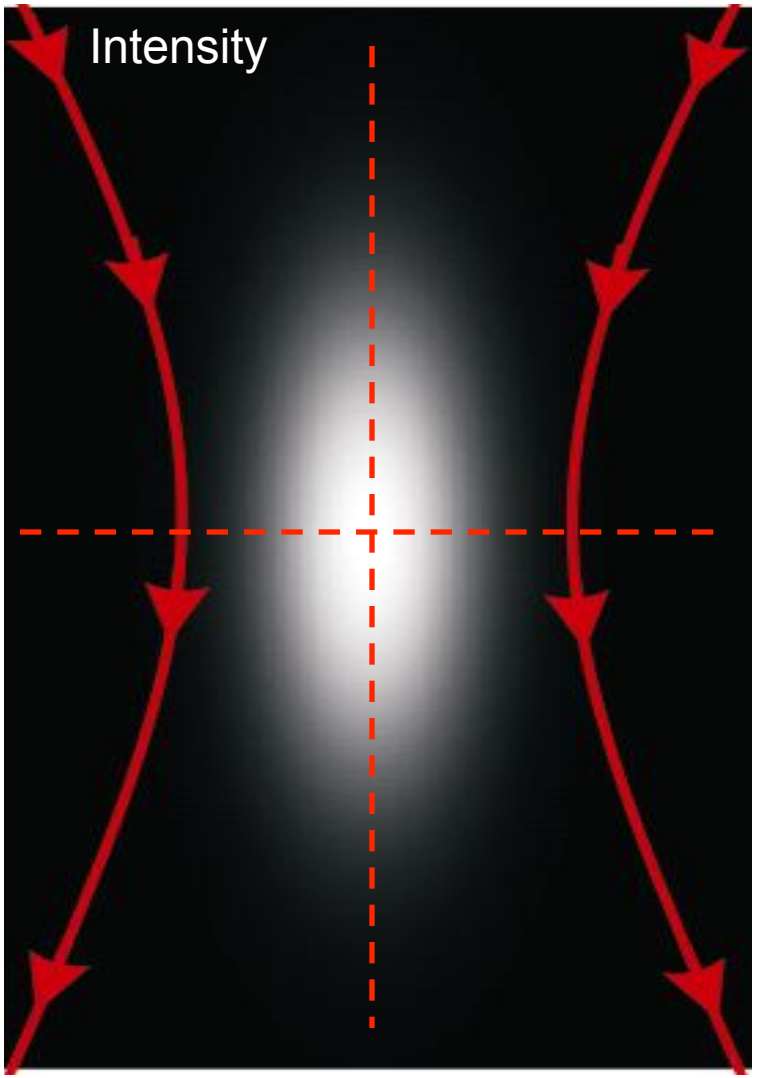
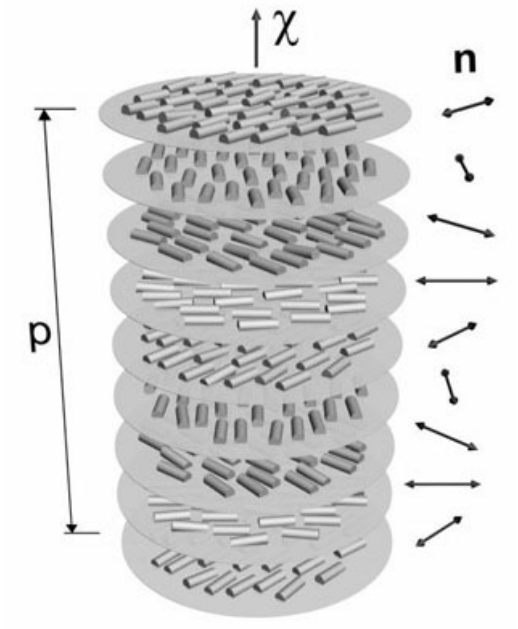
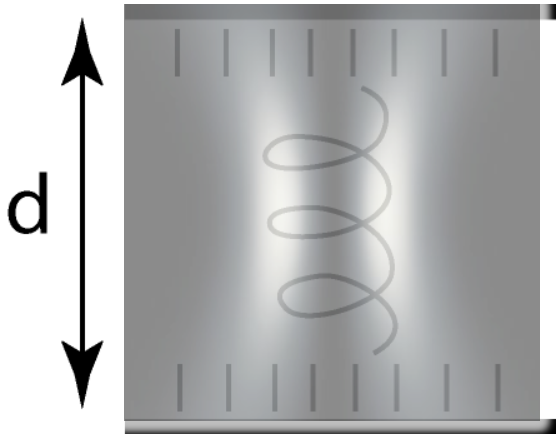
Focused Gaussian beam



$Intensity \sim E^2$

Chiral LCs & vertical surface anchoring





→ Vertical boundary conditions are incompatible with the helical structure

→Result – Frustration & unwinding of the twisted structure

→Control parameter C = d / p

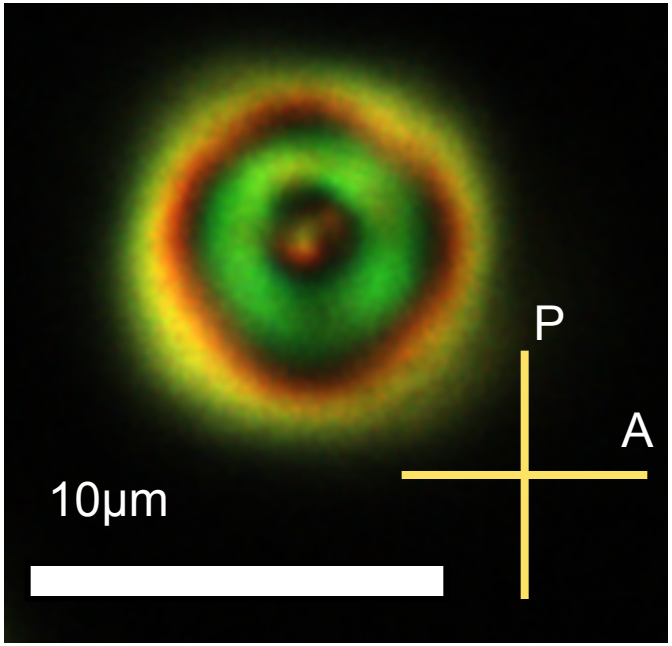
→ External electric field unwinds/winds the structure, depending on field direction;

What is the effect of an LG beam on such a confinement-frustrated unwound LC?

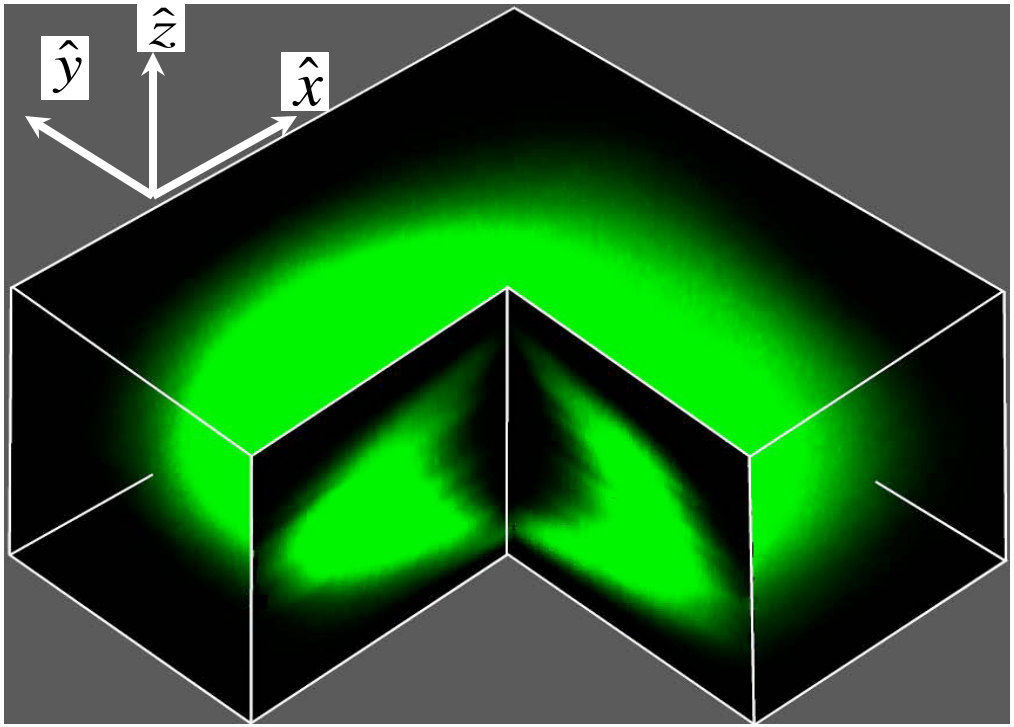
P. Oswald et al., Phys. Rep. 337 (2000)…

Optically-generated stable Toron configuration

2D PM image



3D image

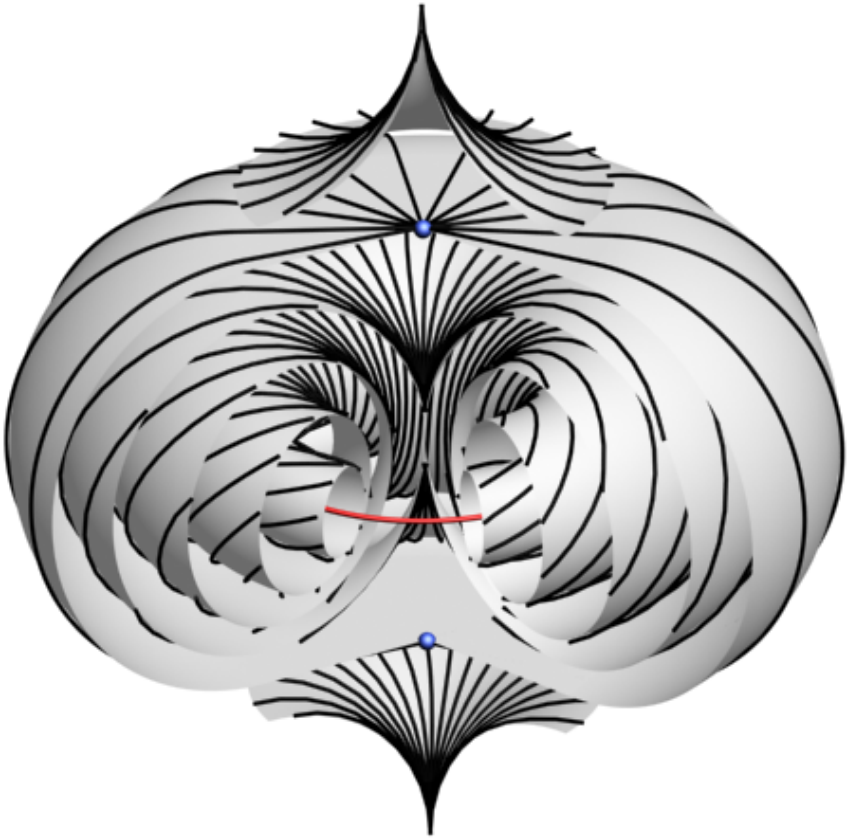


- Shine a 50mW LG beam into the chiral nematic sample for 10-50ms;
- The structure forms spontaneously;
- Long-term stable after switching off the laser light.

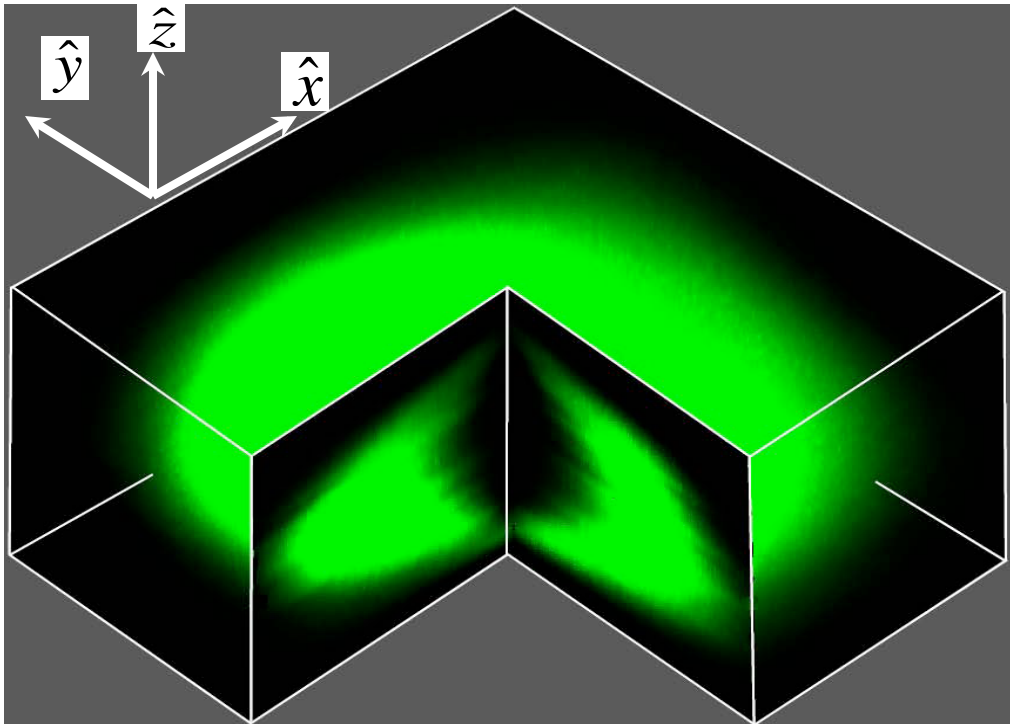
I.I. Smalyukh, Y. Lansac, N. Clark, R. Trivedi, *Nature Materials* **9**, 139-145 (2010).

Optically-generated stable Toron configuration

Reconstructed 3D structure



3D image



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I.I. Smalyukh, Y. Lansac, N. Clark, R. Trivedi, *Nature Materials* **9**, 139-145 (2010).

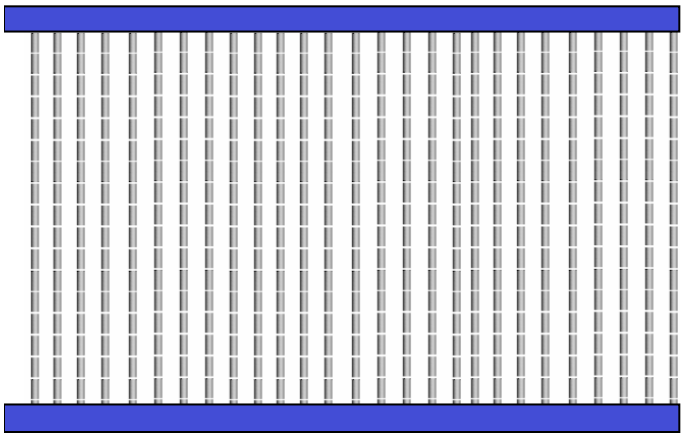
Twist vs. no twist in frustrated cholesteric

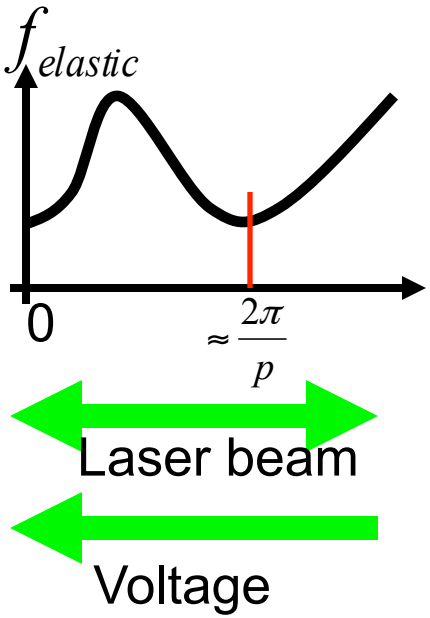
Splay Twist Bend Saddle-splay

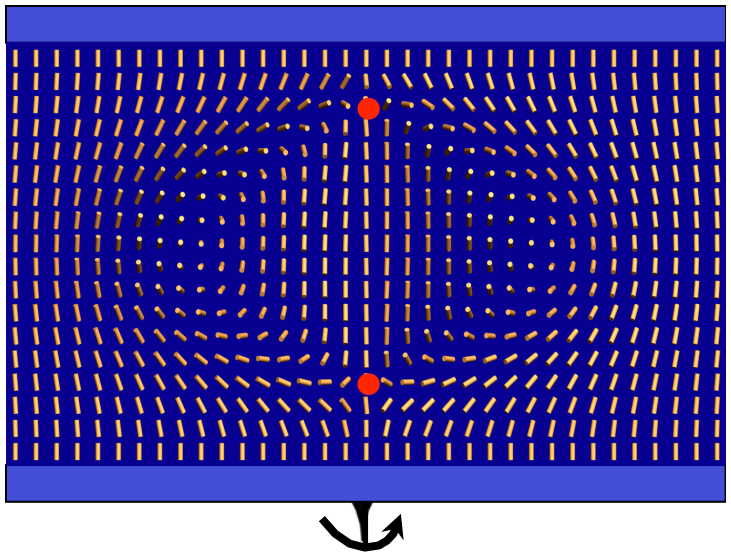
$$f_{elastic} = \frac{K_{11}}{2}(\nabla\cdot\hat{n})^2 + \frac{K_{22}}{2}\left[\hat{n}\cdot(\nabla\times\hat{n}) + \frac{2\pi}{p}\right]^2 + \frac{K_{33}}{2}[\hat{n}\times(\nabla\times\hat{n})]^2 + f_{24}$$

$$f_{24} = -K_{24}\{\nabla\cdot[\hat{n}(\nabla\cdot\hat{n}) + \hat{n}\times(\nabla\times\hat{n})]\}$$

Untwisted cholesteric cell



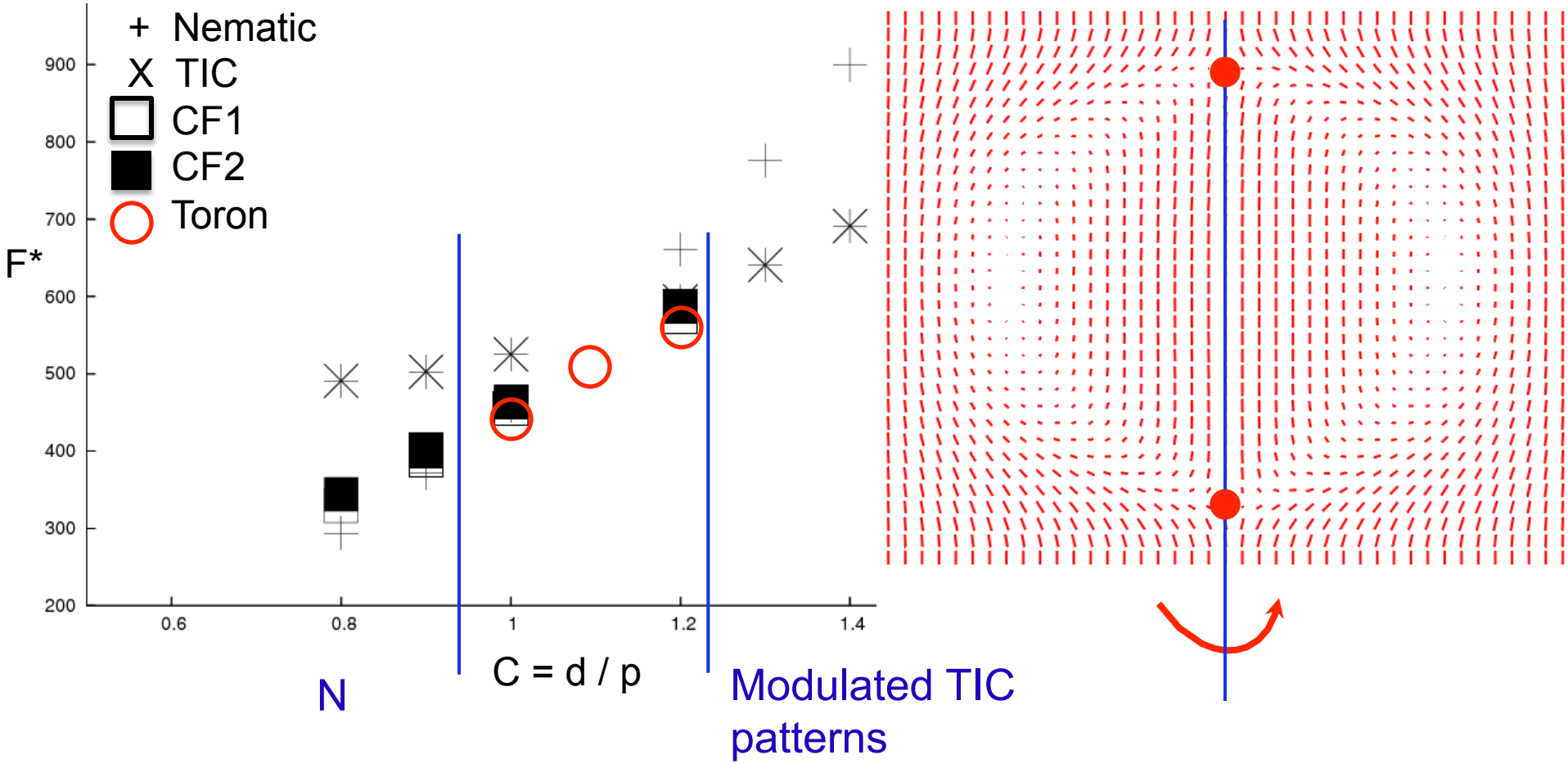




→ Cholesteric pitch *p*~*d*;
→ Strong energetic barrier between the structures >> $K_B T$;
→ Both states can be stable for long time;
→ Switching by a focused laser beam or applying voltage;

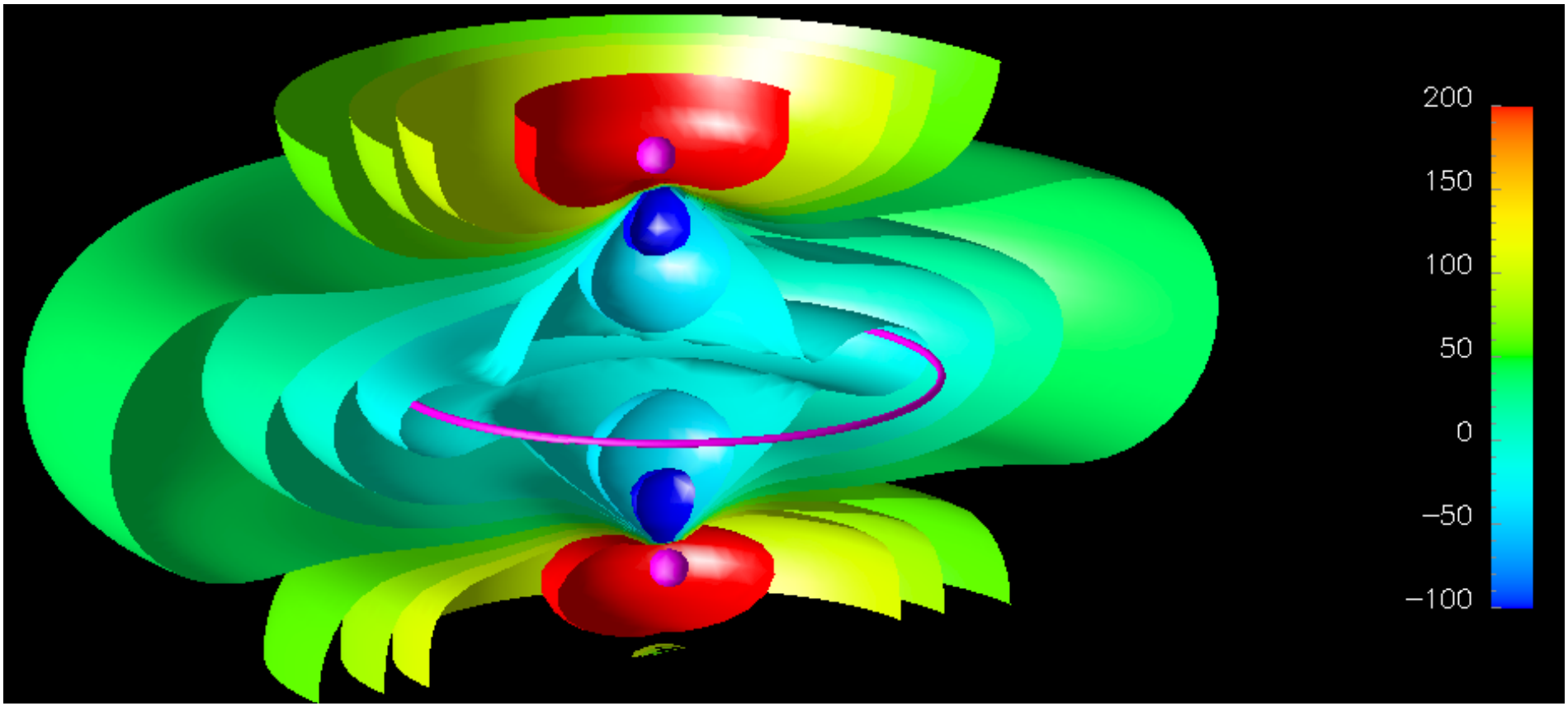
Free energy of localized LC structures

Axially-symmetric localized **n**(**r**) minimizing elastic energy



I.I. Smalyukh, Y. Lansac, N. Clark, R. Trivedi, *Nature Materials* **9**, 139-145 (2010).

Total free energy density isosurfaces



- The energy is minimized in the double-twisted part of the structure
- Point defects – energetically-costly part of the structure;
- Total free energy slightly lower than that of the unwound state;

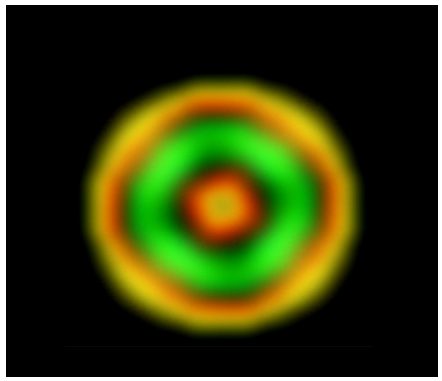
I.I. Smalyukh, Y. Lansac, N. Clark, R. Trivedi, *Nature Materials* **9**, 139-145 (2010).

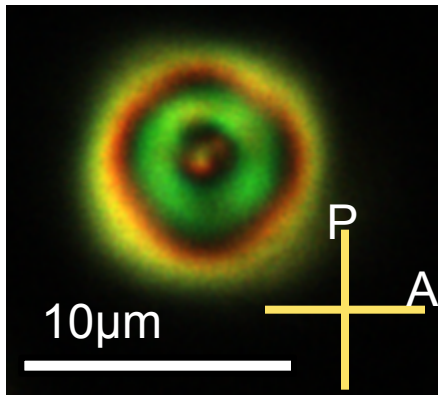
Computer simulations vs. experiments

Simulated

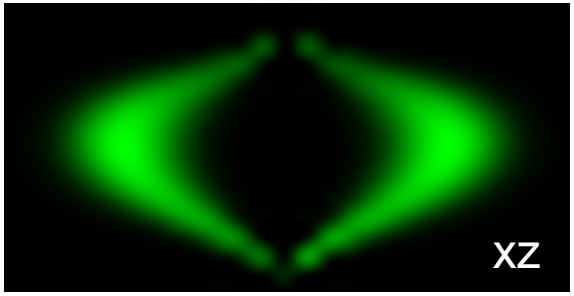
Experimental

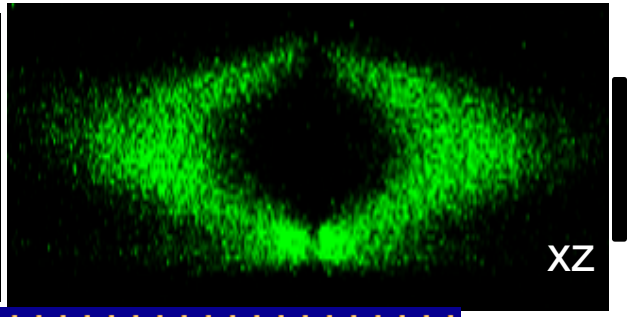
Polarizing microscopy



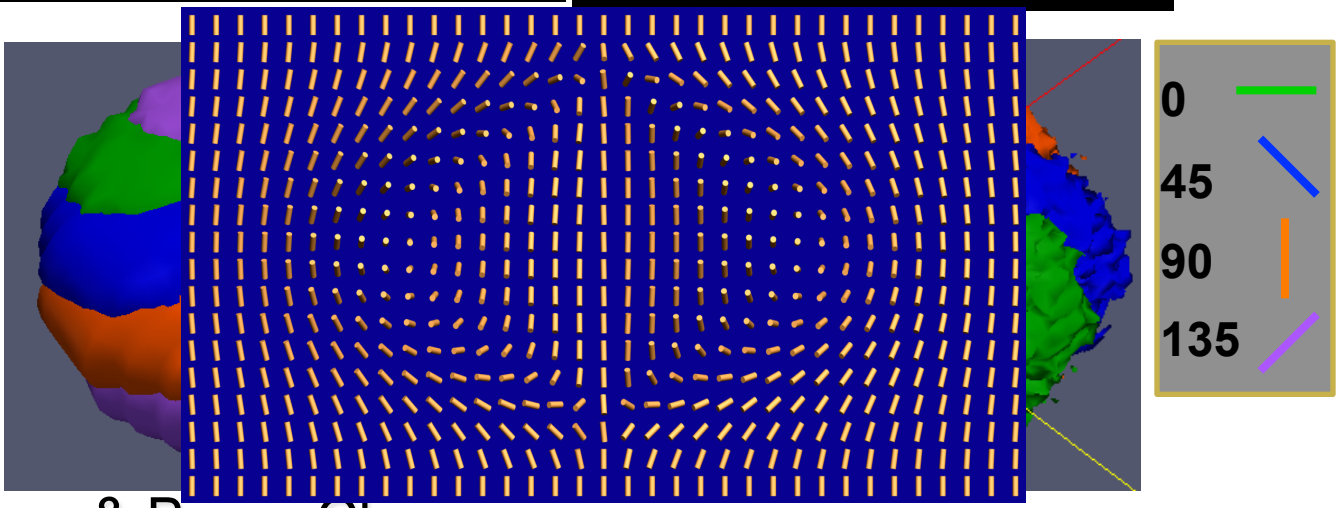


3-photon excitation fluorescence polarizing m-py (3PEF-PM)





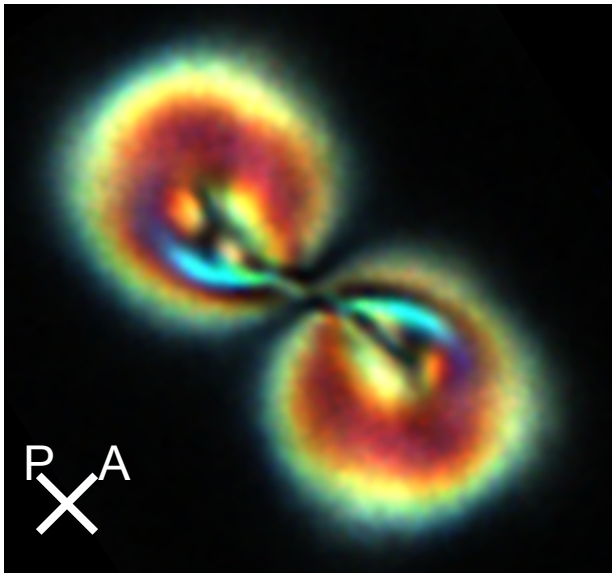
Paraview presentation of 3D images (3PEF-PM)

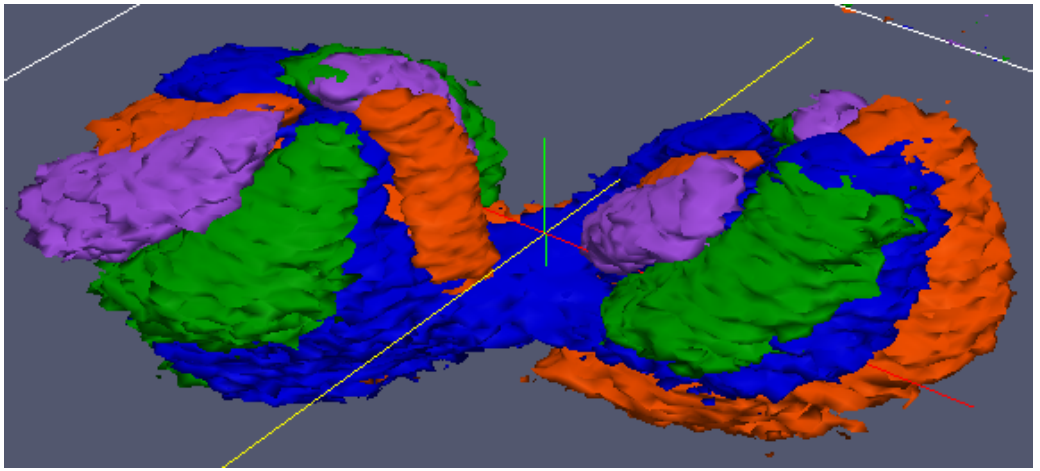


- Paul Ackerman & Bryan Chen

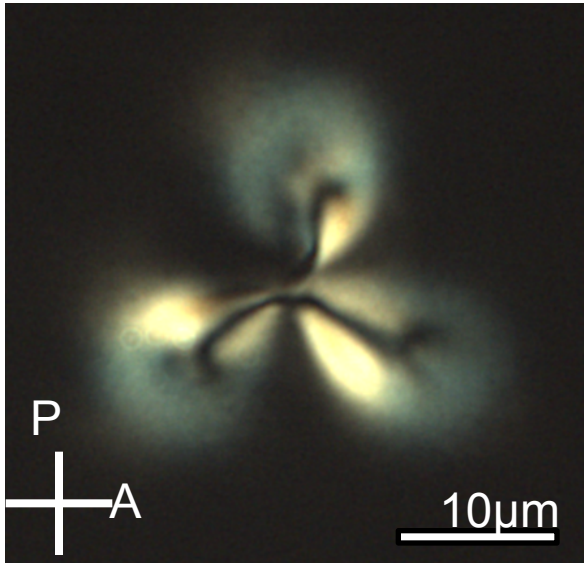
Inter-linked Torons: Duo & Trio

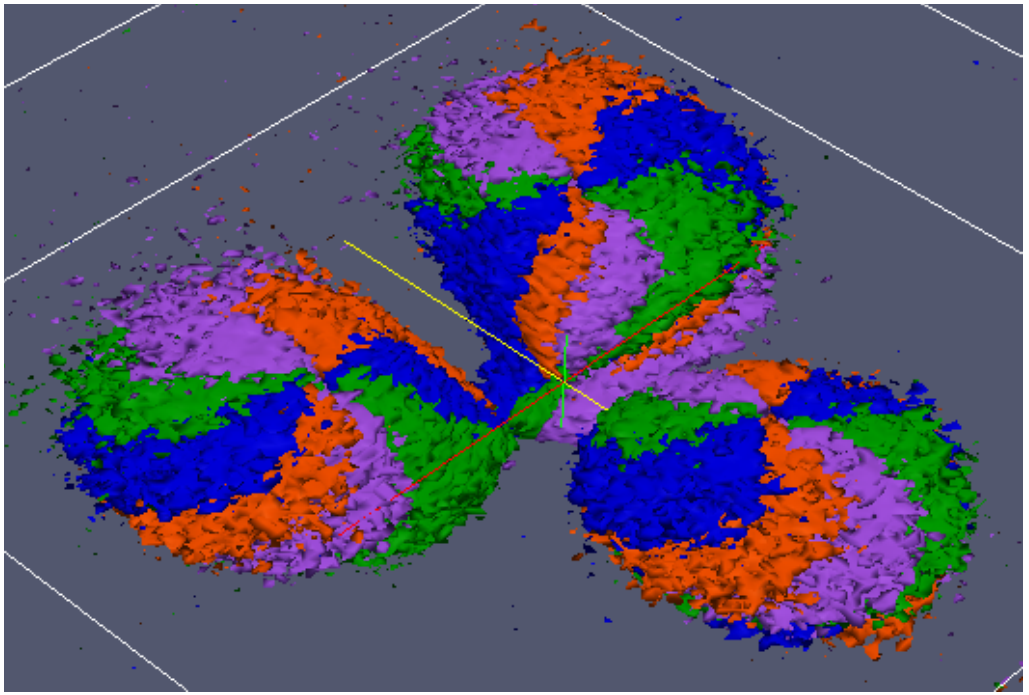
Two linked torons

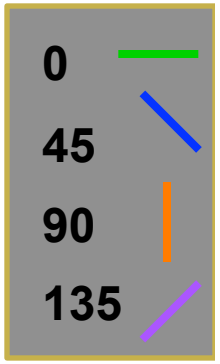




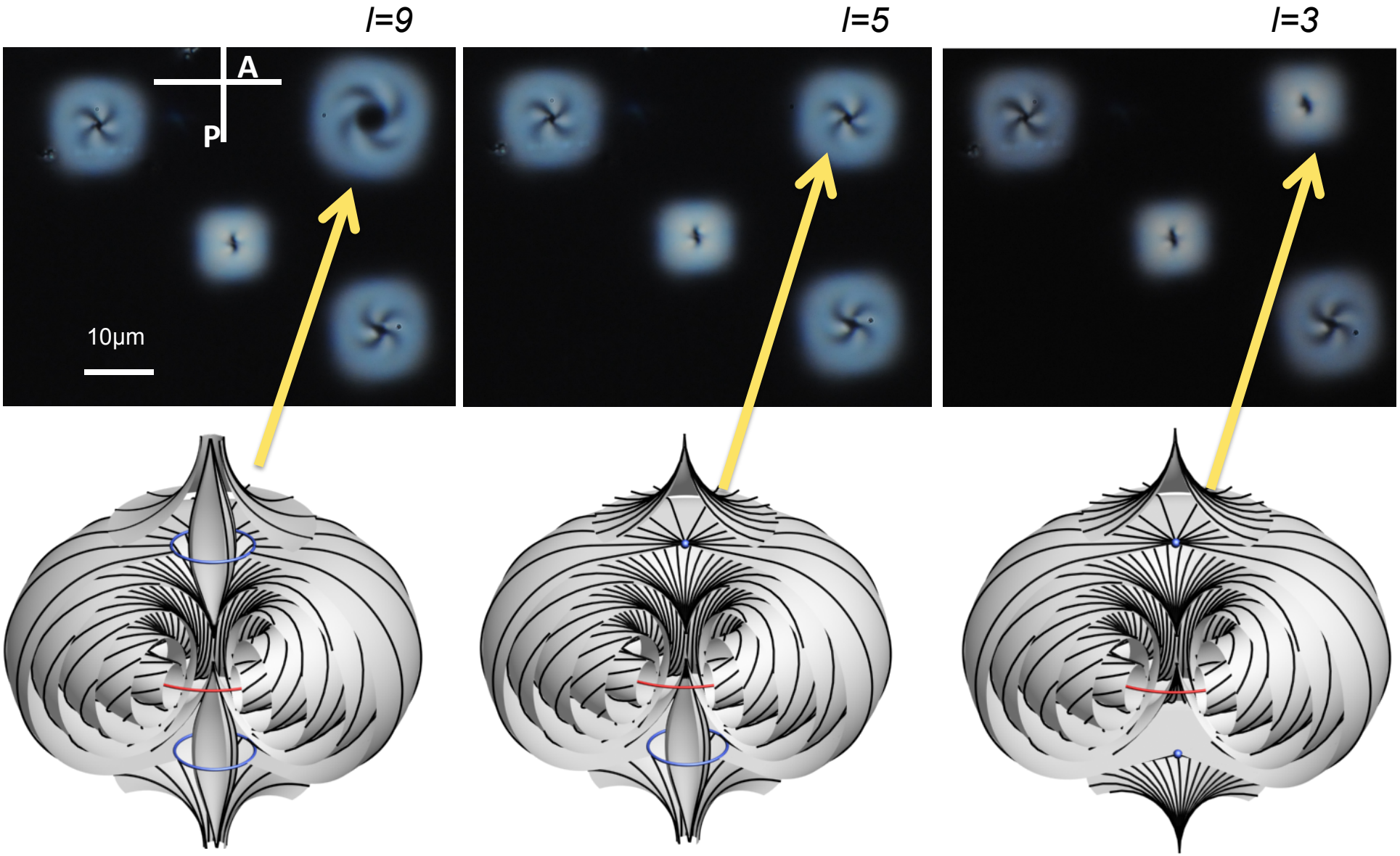
Three linked torons





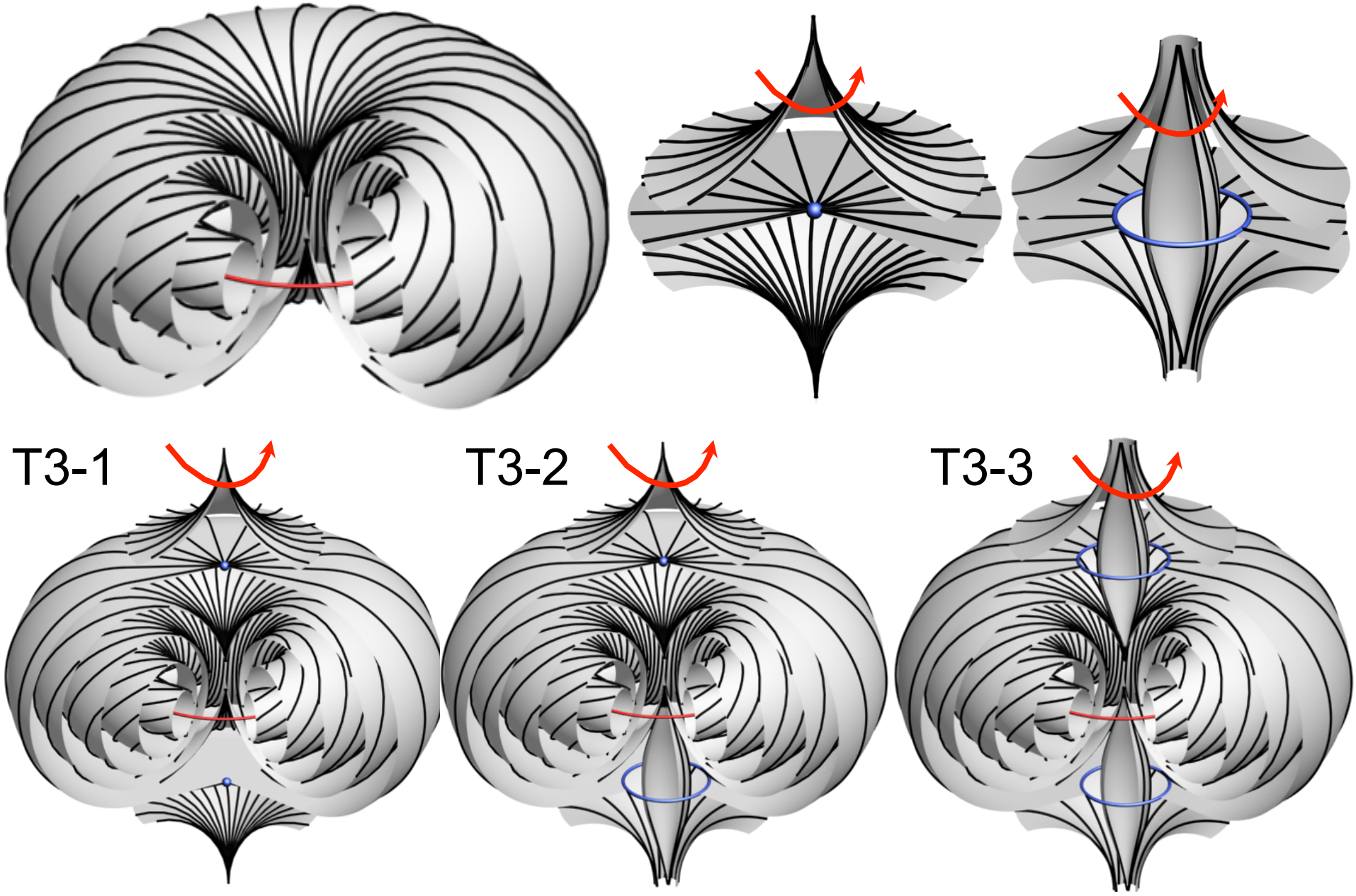


Optical control of defects in Torons

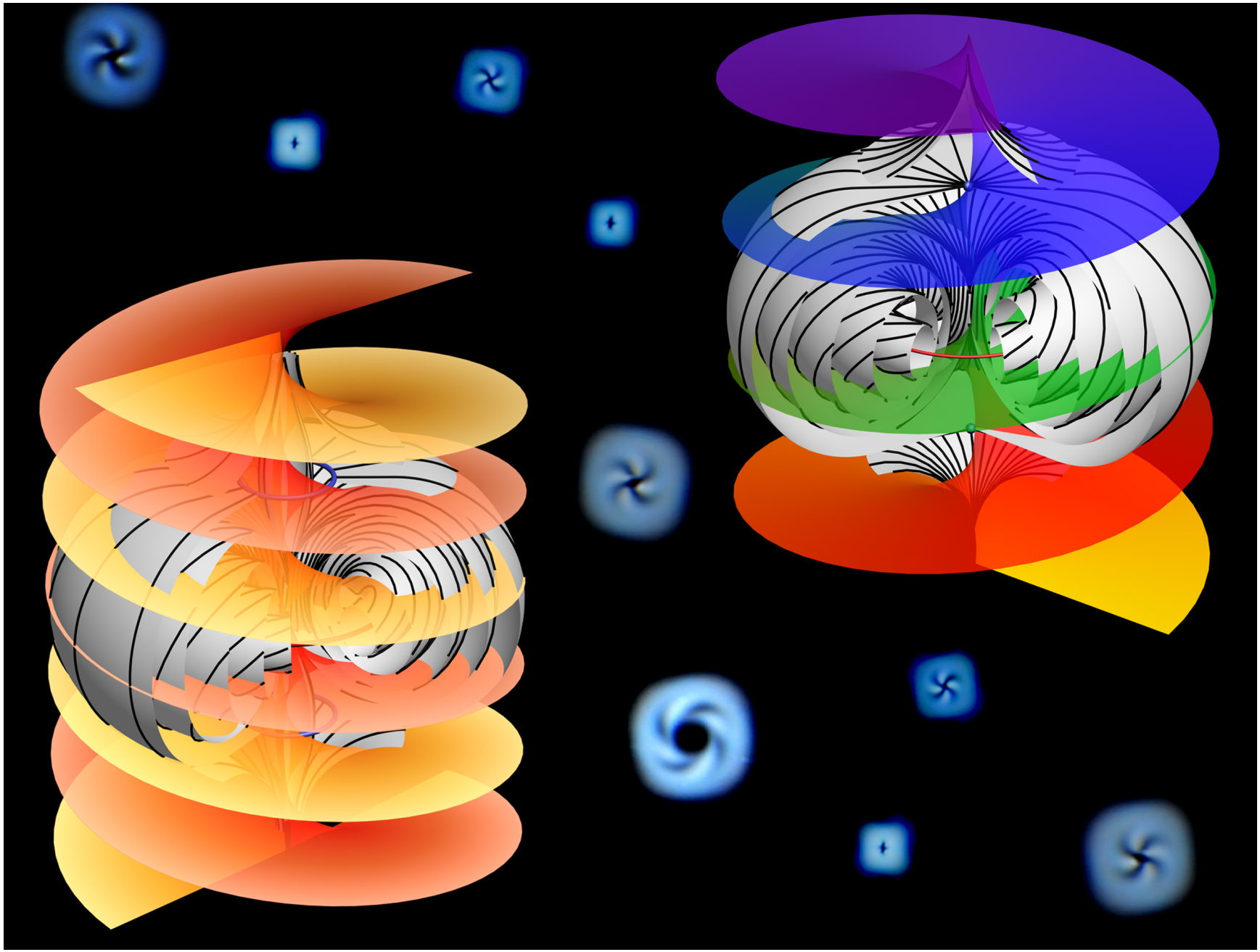


I.I. Smalyukh, Y. Lansac, N. Clark, R. Trivedi, *Nature Materials* **9**, 139-145 (2010).

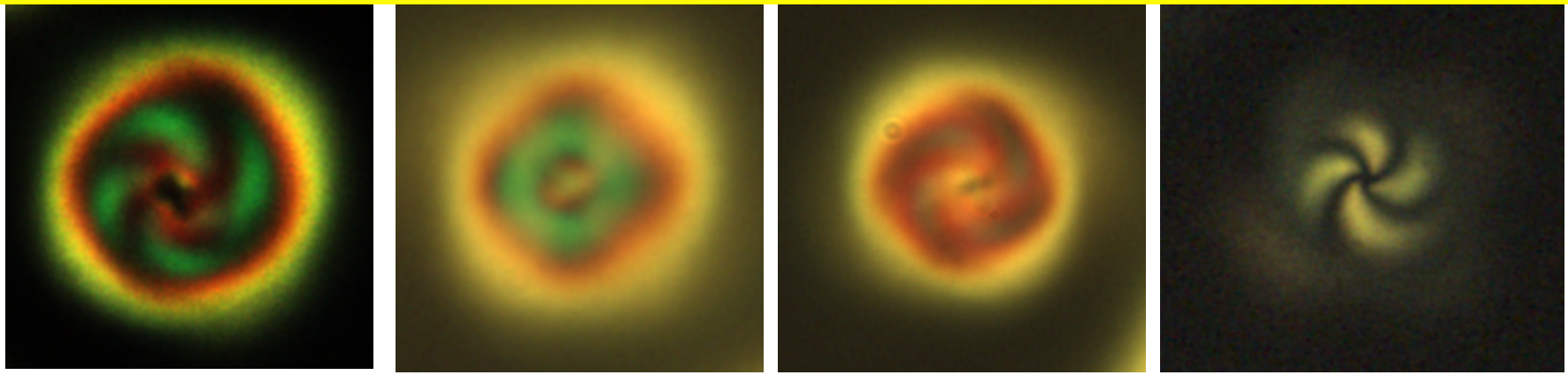
Types of LG-beam-generated Torons

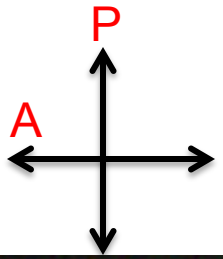


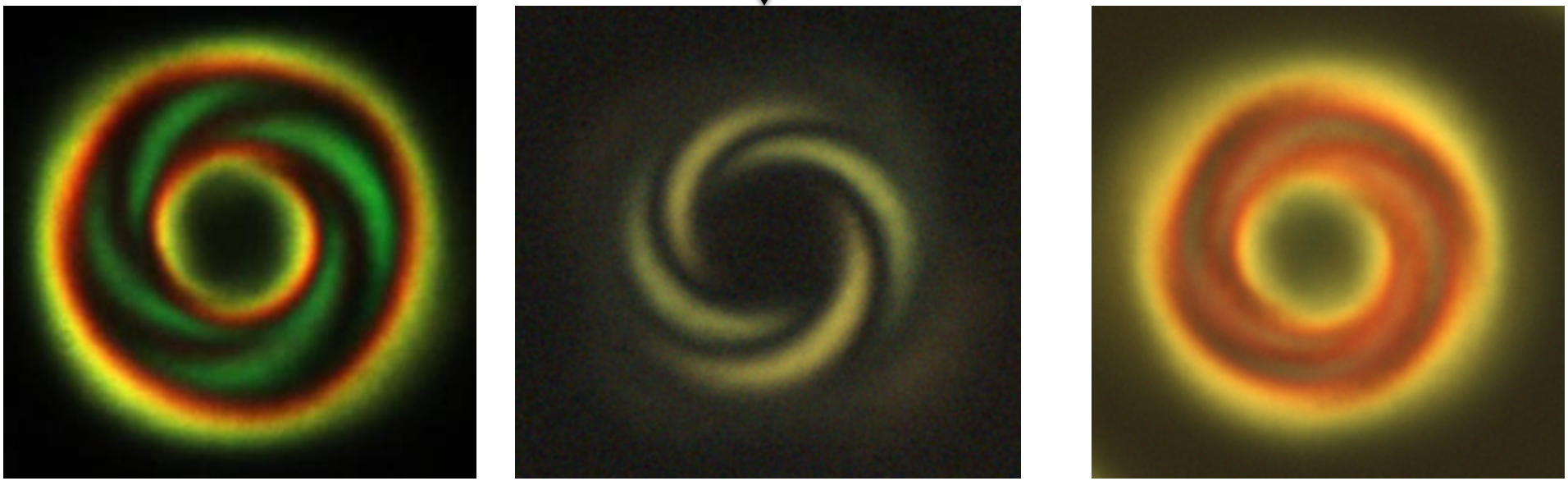
Control of LC defects using optical singularities



Examples of localized structures we generate

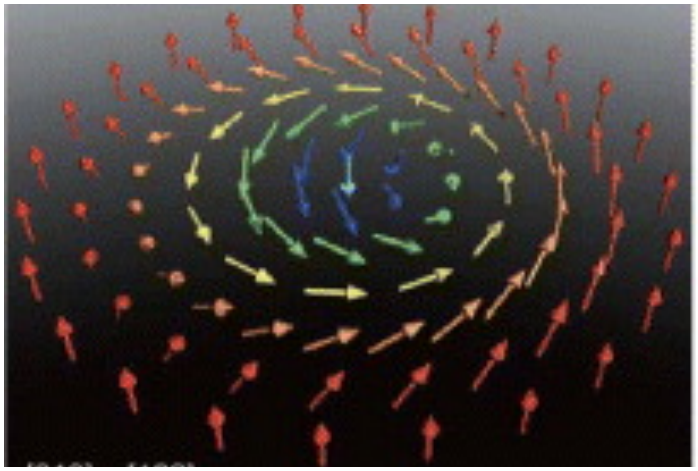


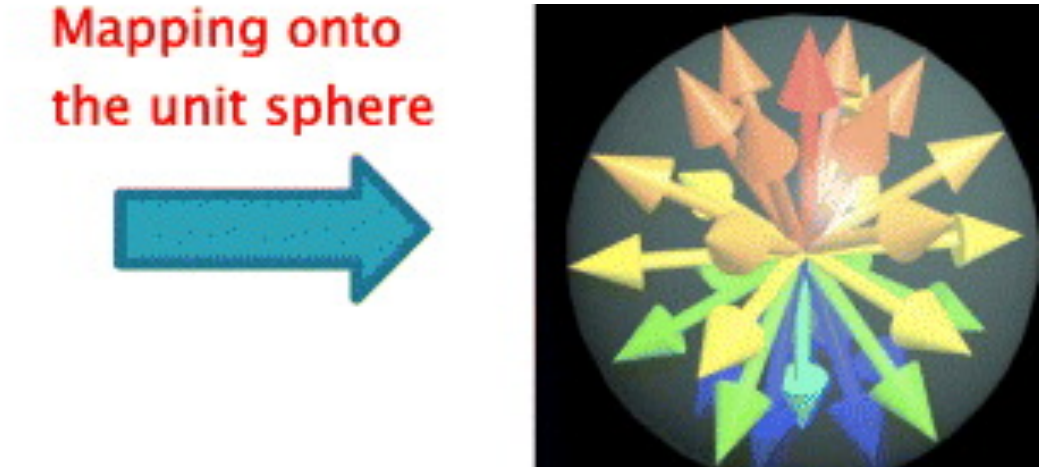




Nonsingular configurations: Skyrmions, Baby Skyrmions, Merons, Bimerons…

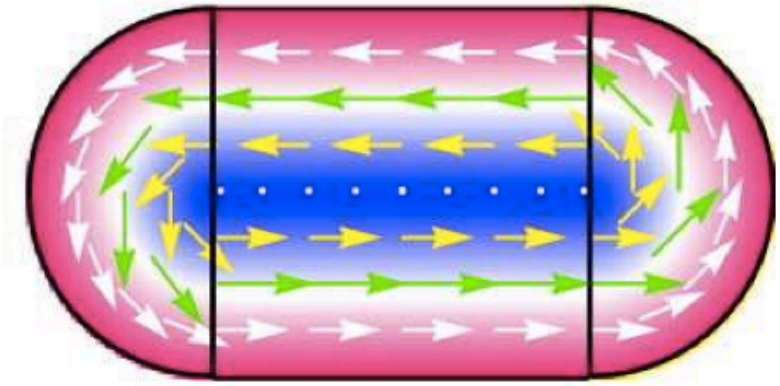
Skyrmion



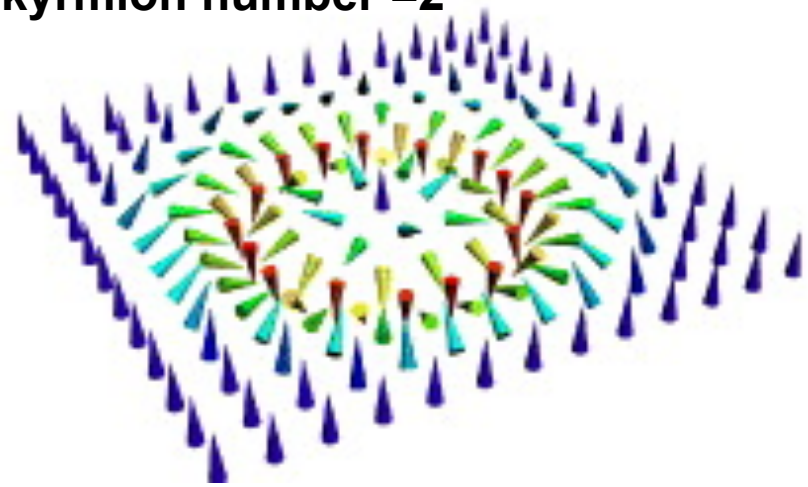


Skyrmion number - how many times the mapping wraps the unit sphere.

Bimeron (two half-disc domains)



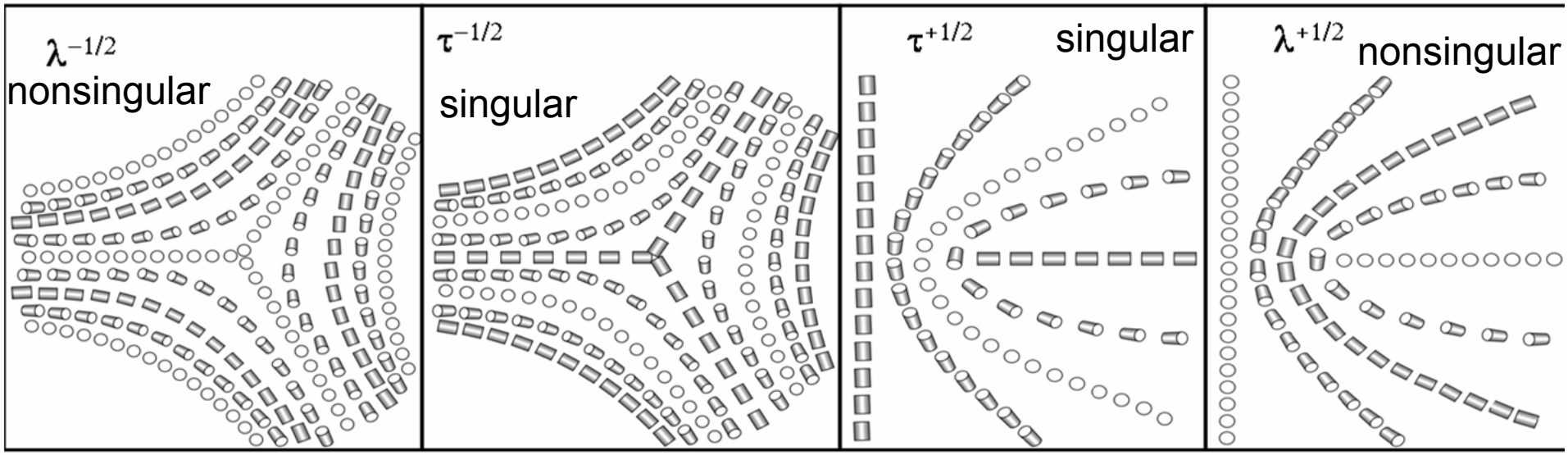
Skyrmion number =2



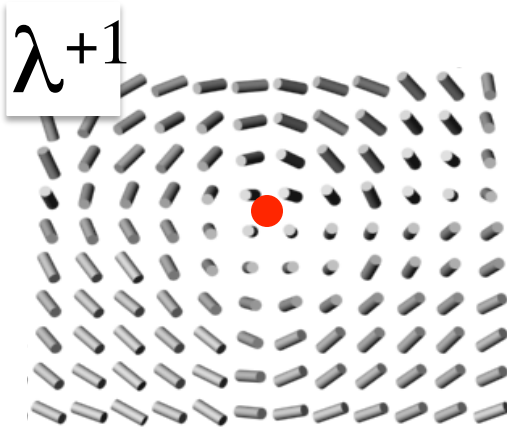
Nagaosa & Tokura (2012)

http://park.itc.u-tokyo.ac.jp/nagaosa-lab/

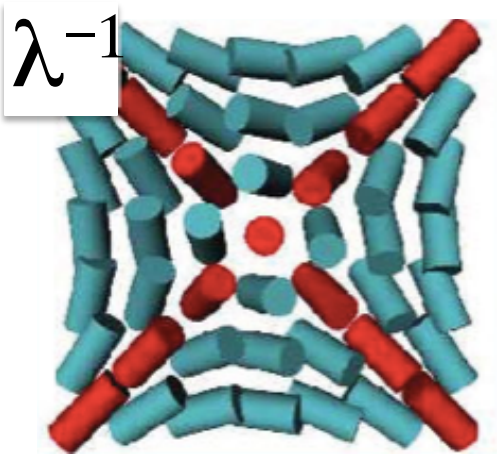
Wedge & twist disclinations in cholesterics



Twist-escaped non-singular defect lines

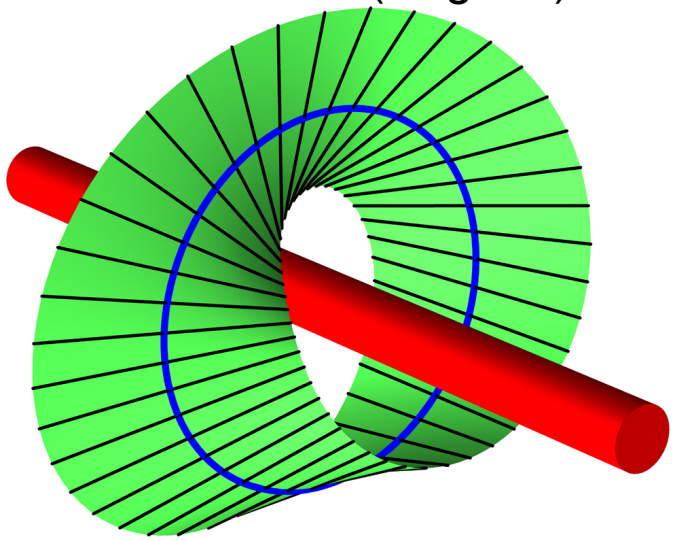


Trushkevych, et al. *APL* **97**, 201906 (2010).



Tkalec et al, PRL **103**, 127801 (2009)

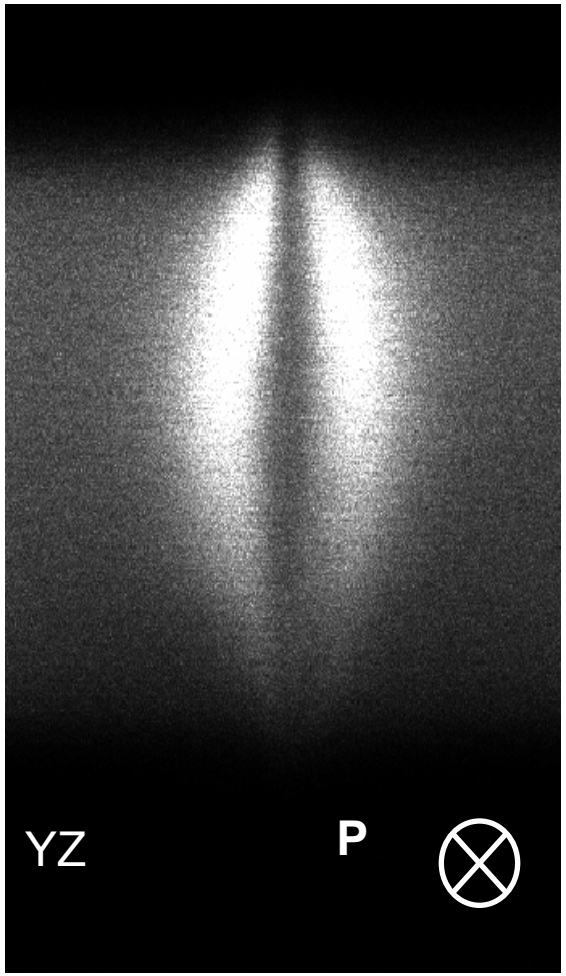
Twist disclination (singular)

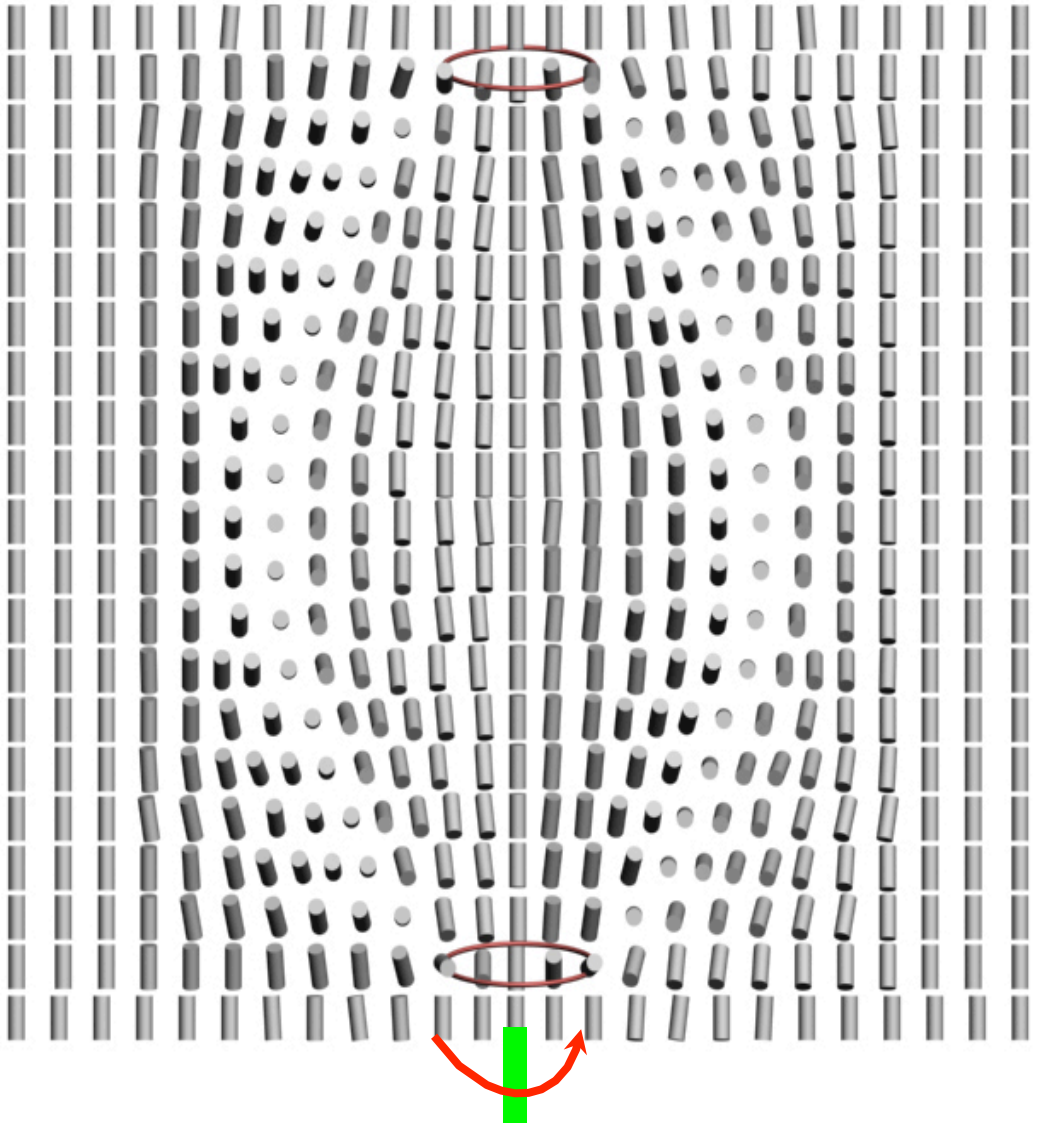


Smalyukh et al., Phys. Rev. E **72**, 061707 (2005).

“Baby Skyrmion” in a confined cell geometry

Vertical cross-section

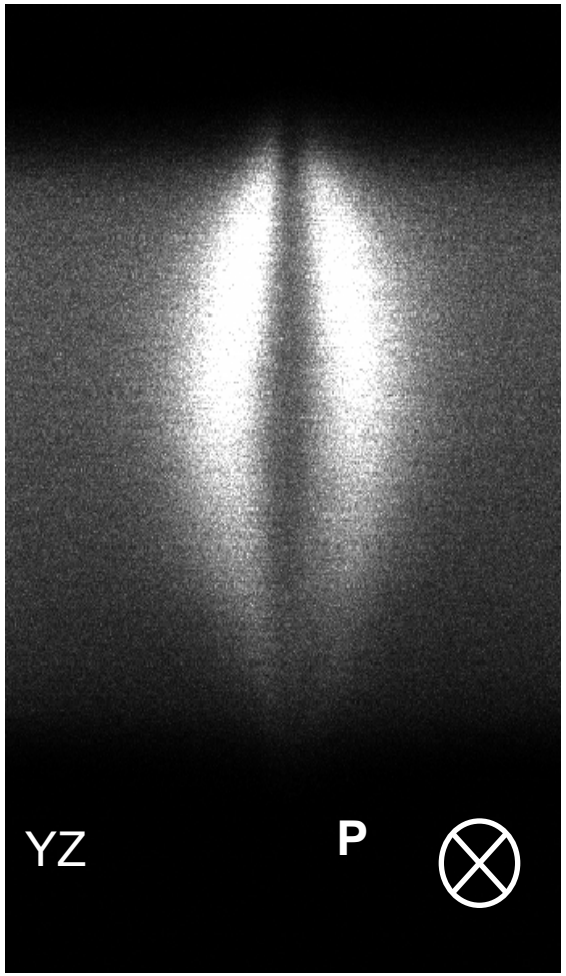


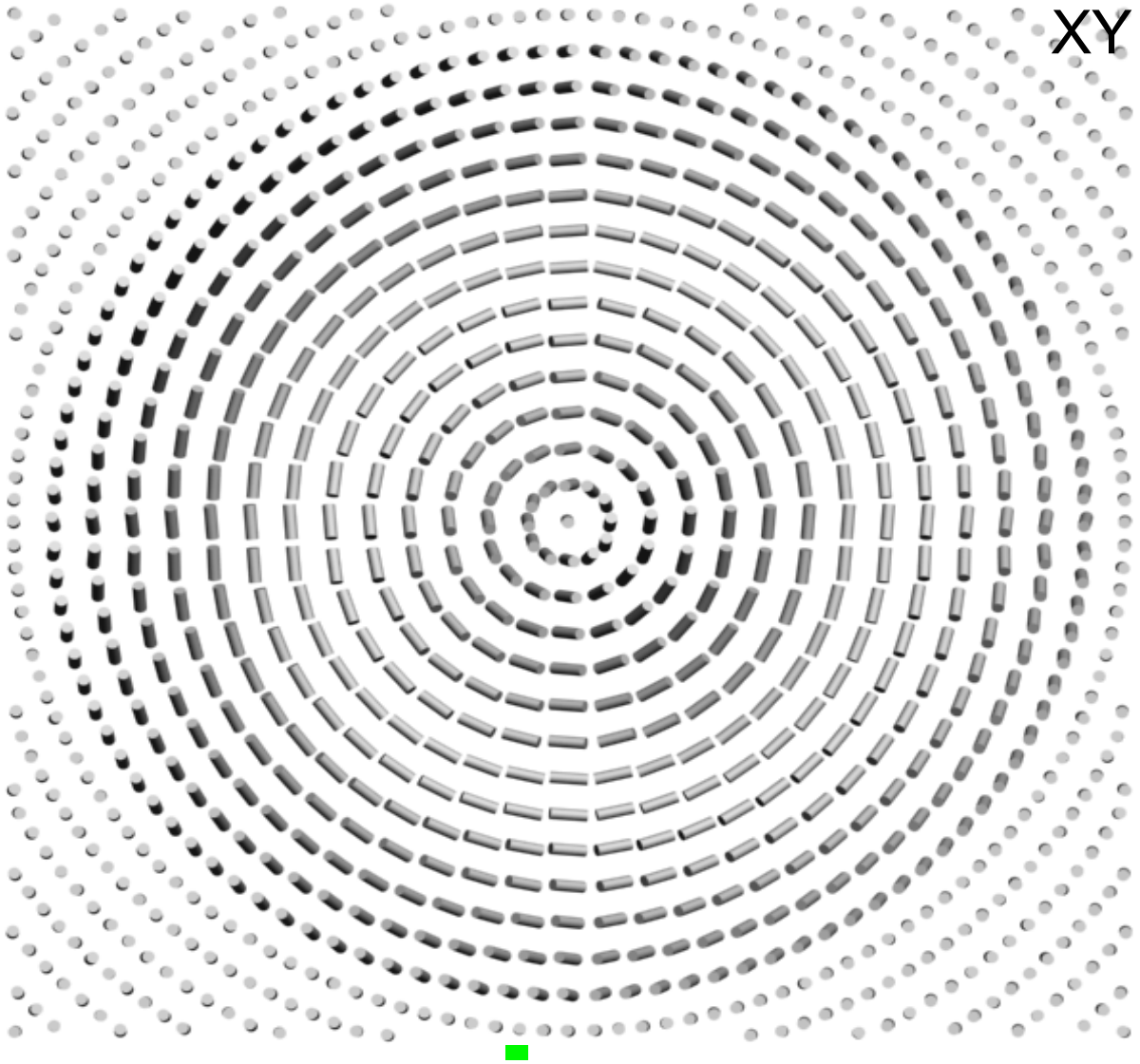


- Double-twist cylinder capped by twist disclinations

“Baby Skyrmion” in a confined cell geometry

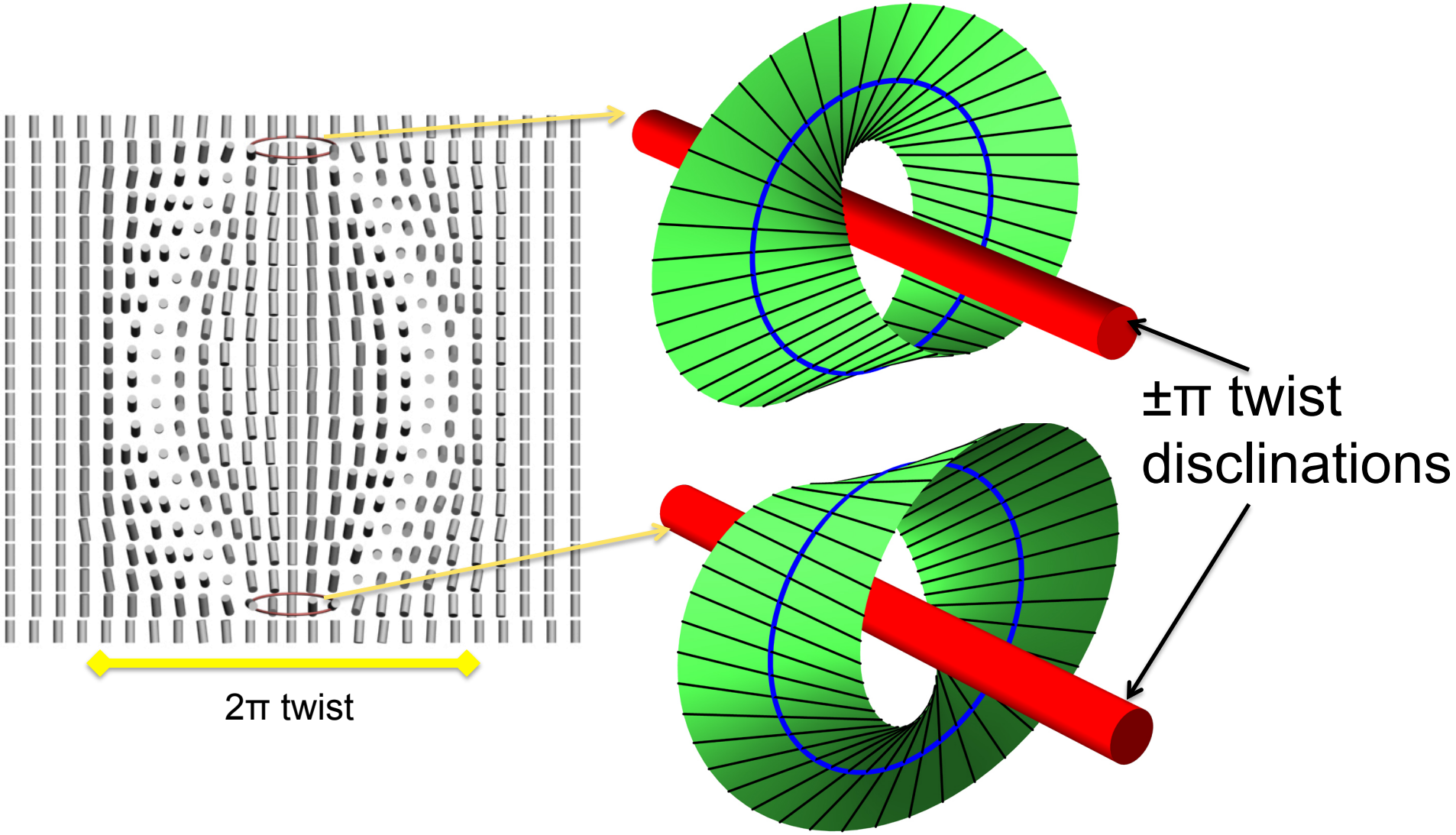
Vertical cross-section



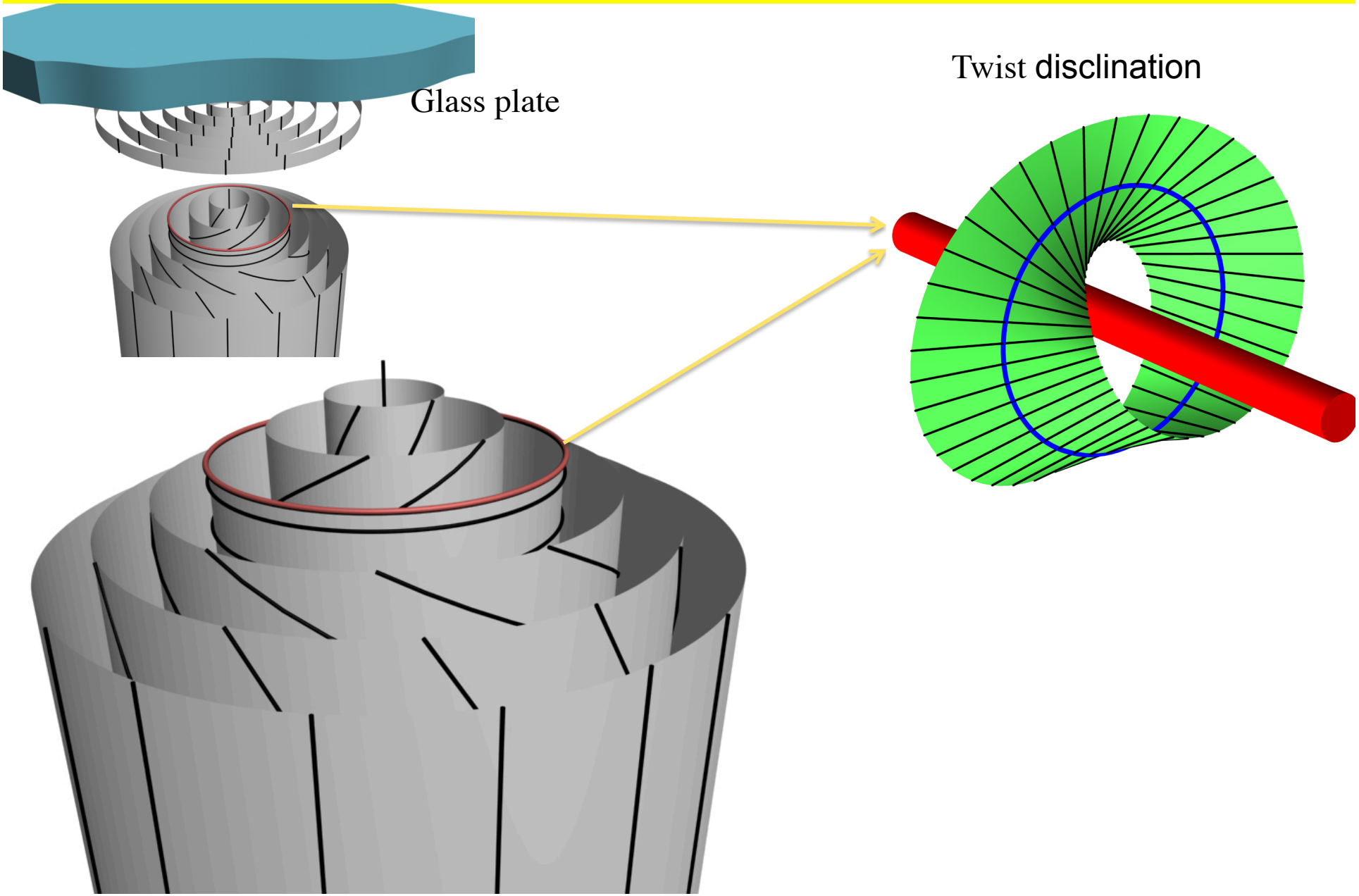


- Double-twist cylinder capped by twist disclinations

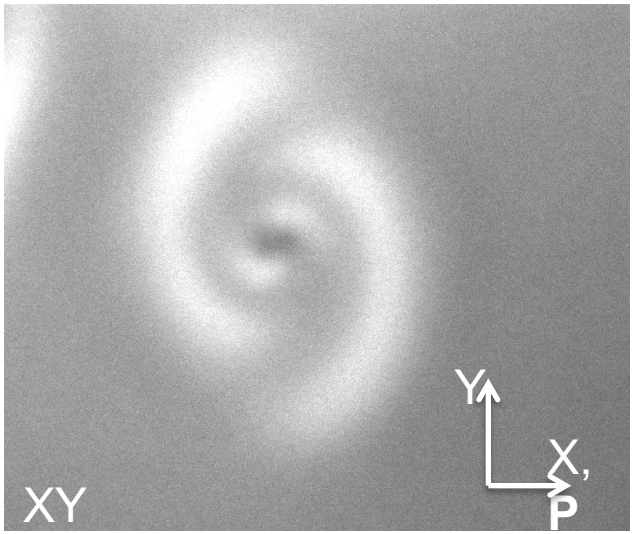
“Baby Skyrmion” in a confined cell geometry

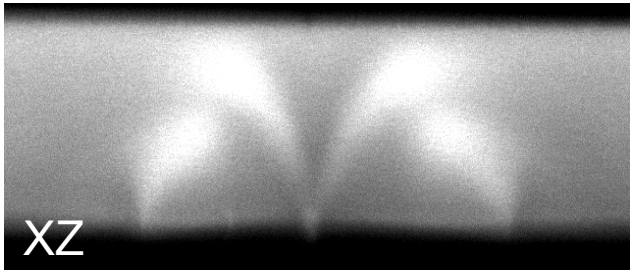


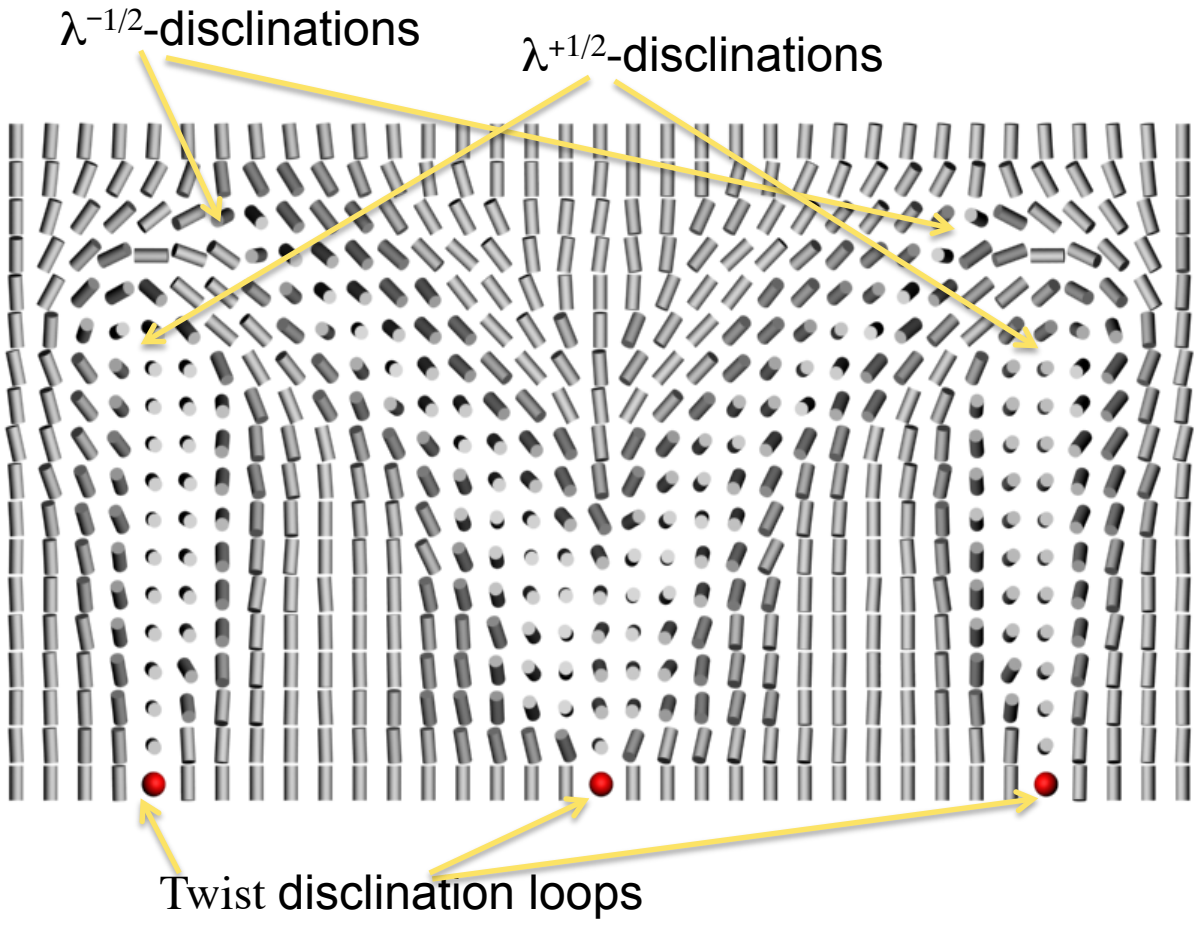
“Baby Skyrmion” in a confined cell geometry



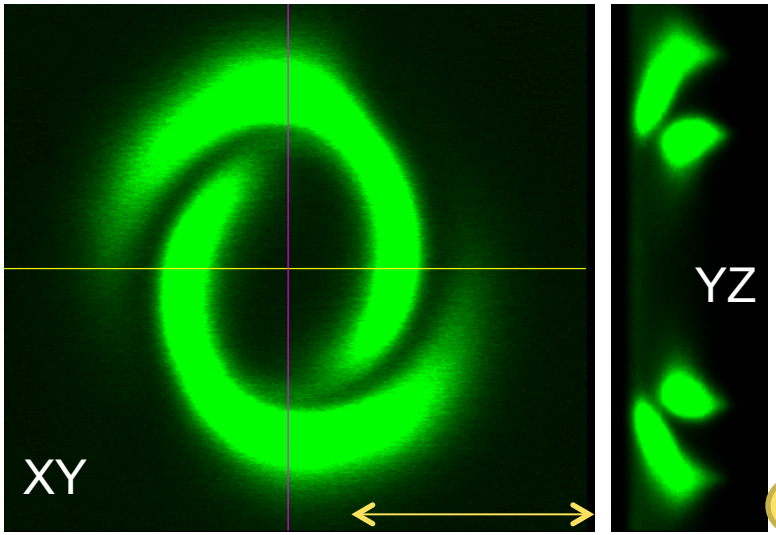
Other twist-stabilized localized configurations

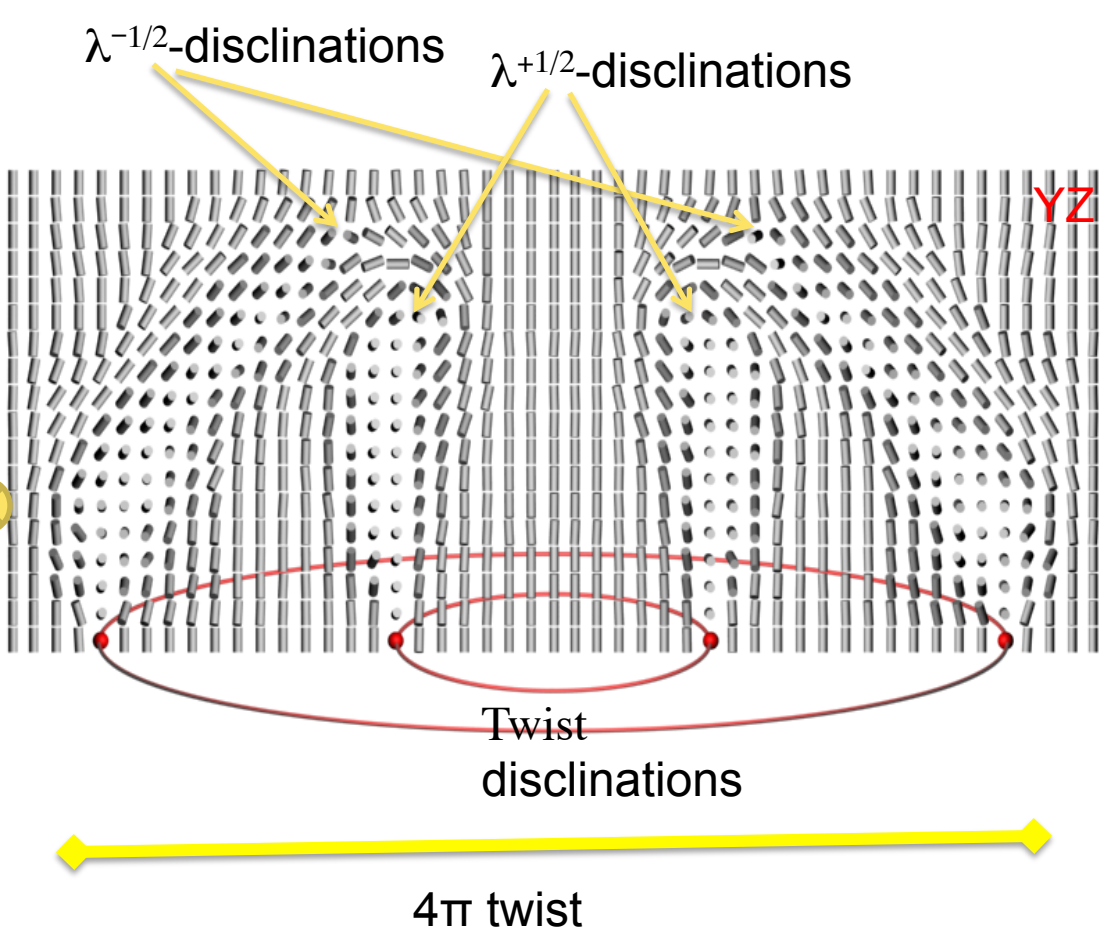






Other twist-stabilized localized configurations

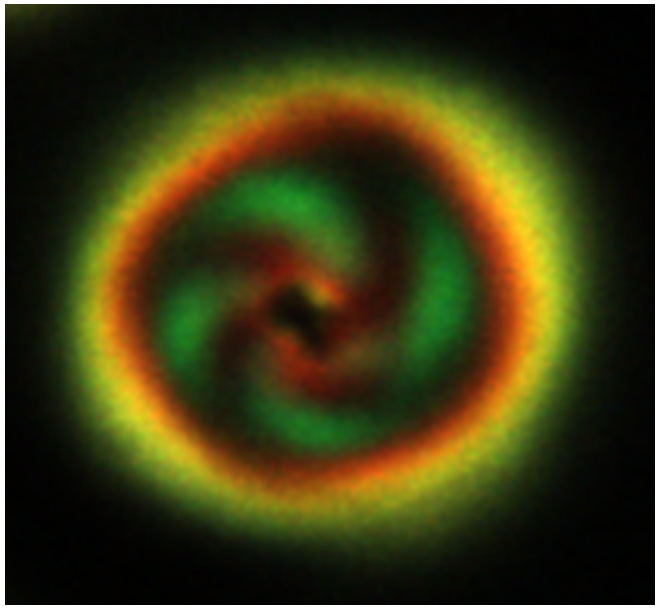


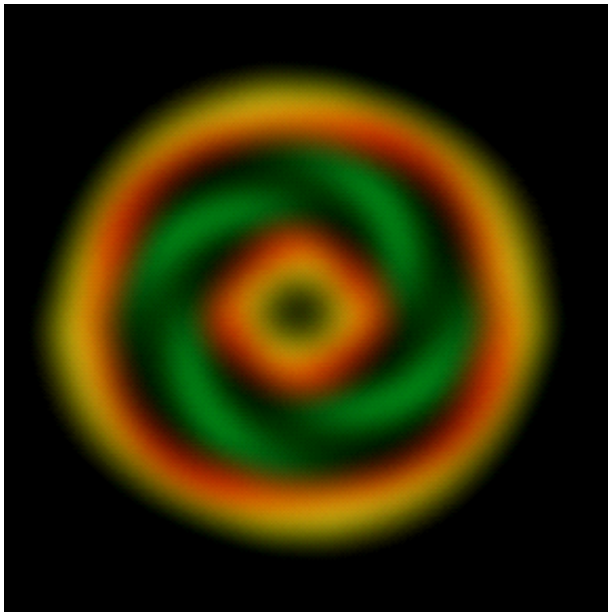


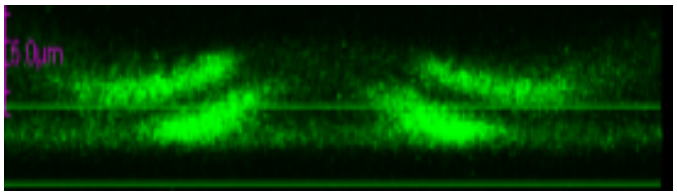
Other twist-stabilized localized configurations

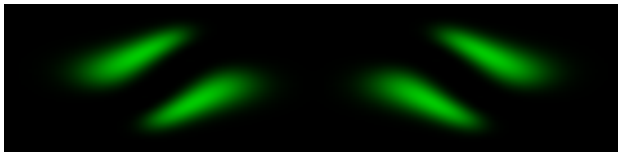
Experimental

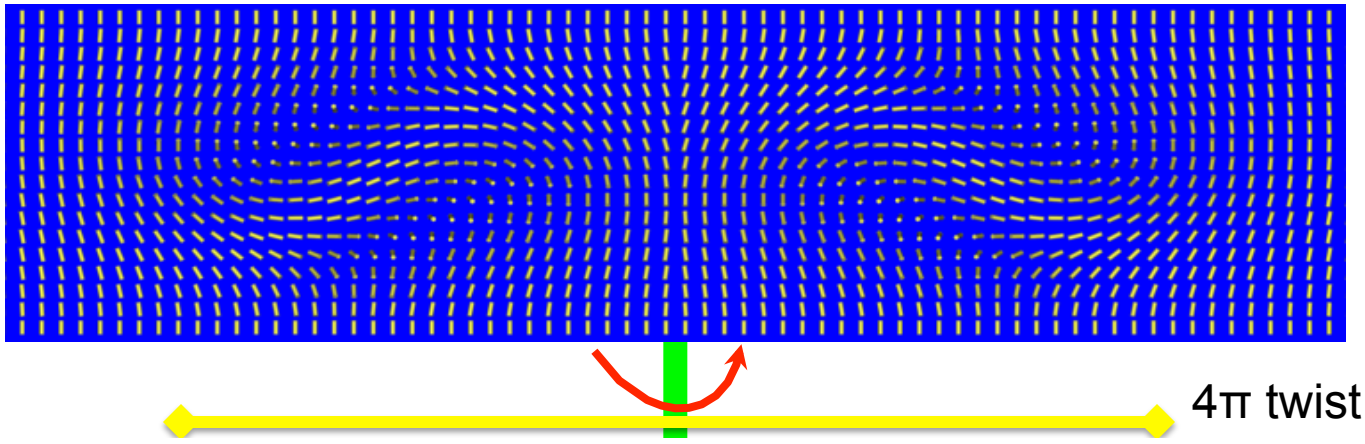
Simulated







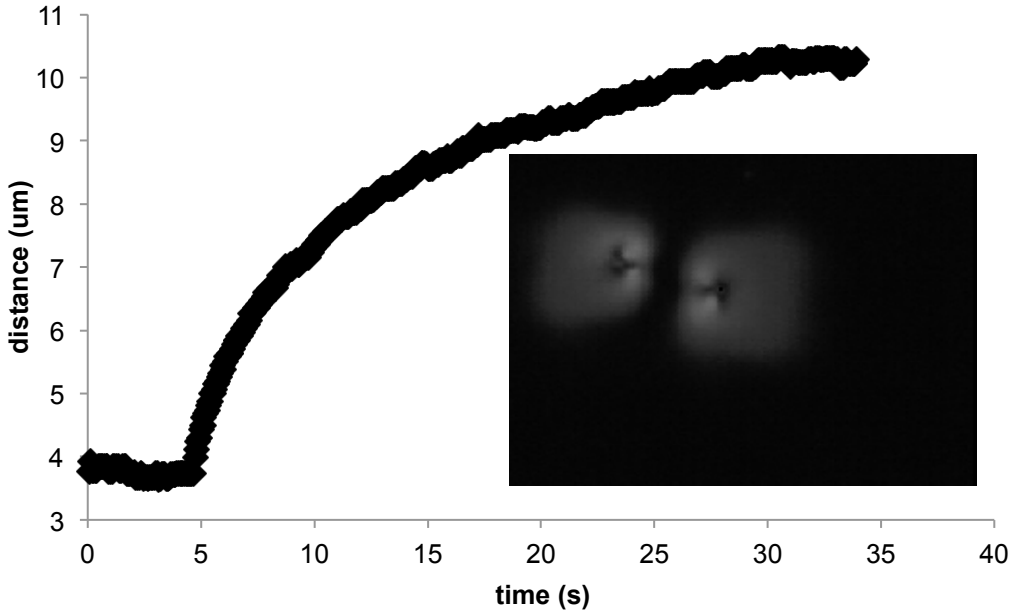


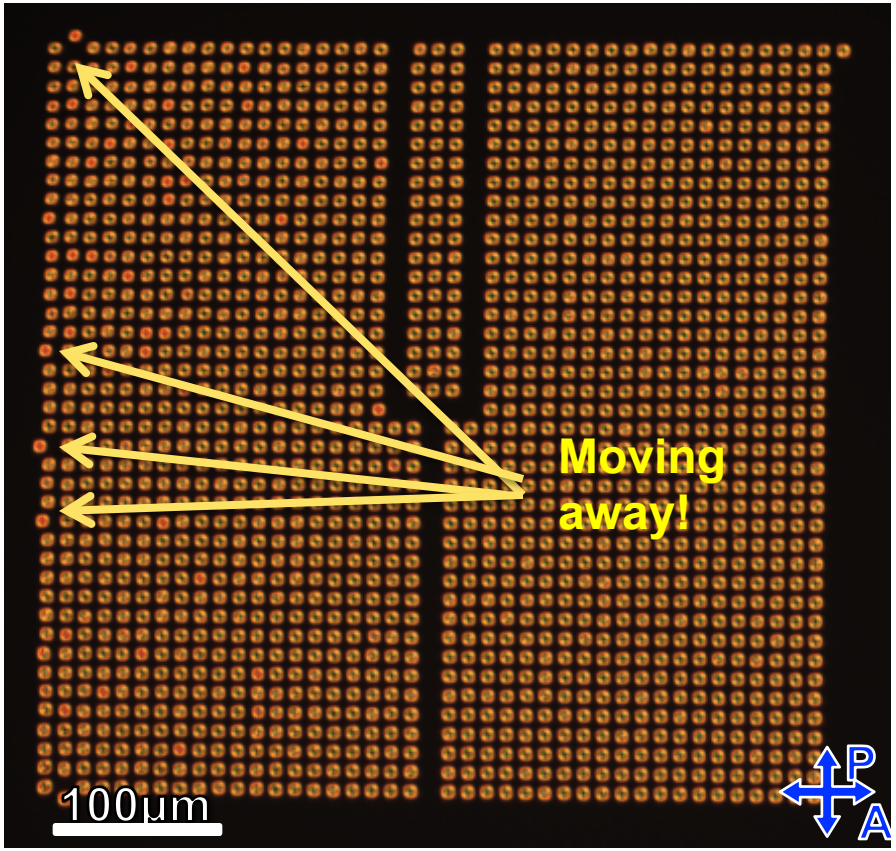


Nonsingular director configuration

Modes of generation: sticking & mobile

Generated using laser powers just above threshold:
→Short-range repulsion;
→Brownian motion





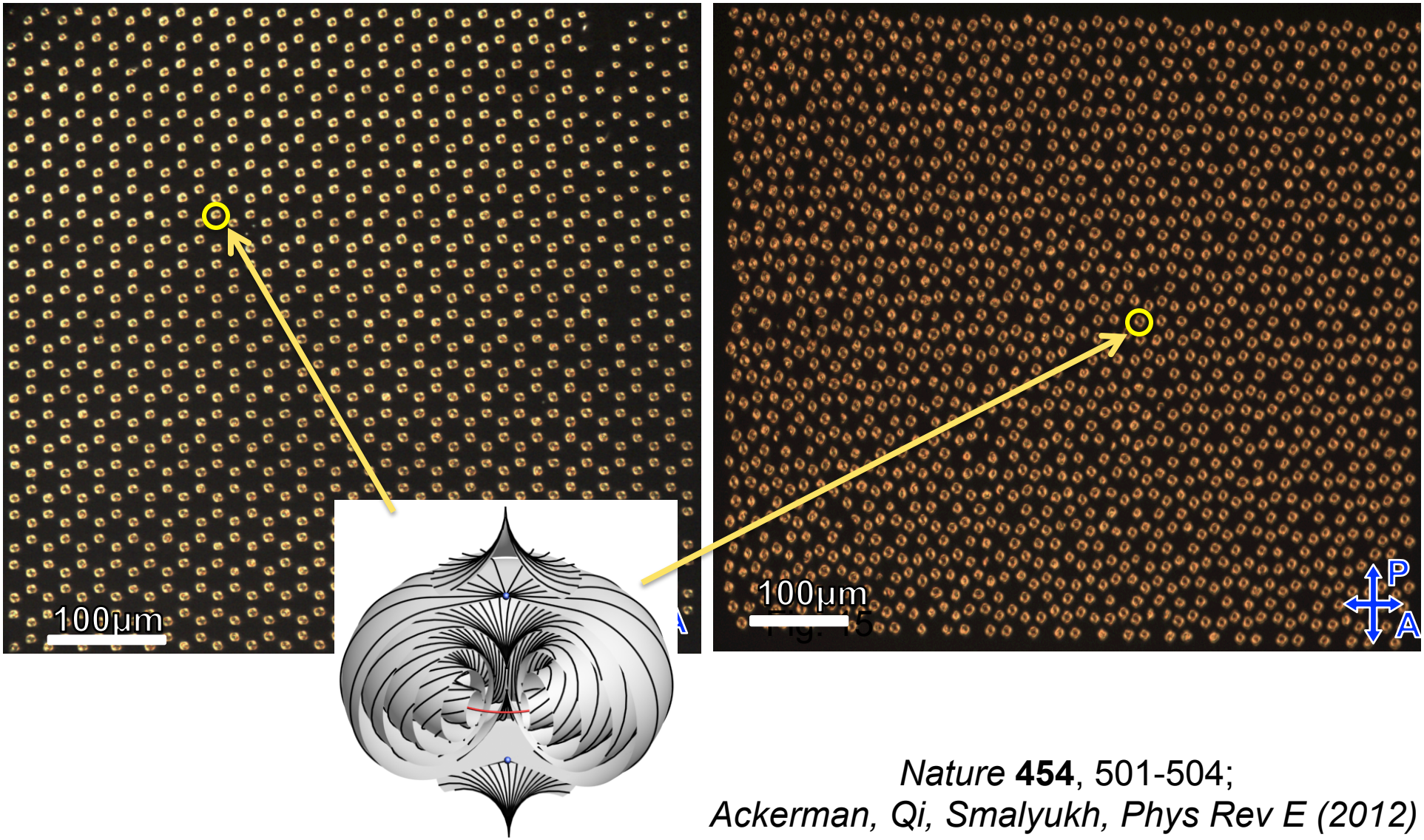
Generated with laser powers >70mW:
→Pinned in the location of generation;
→No Brownian motion;
→No repulsive interactions

Ackerman et al, Phys Rev E (2012)

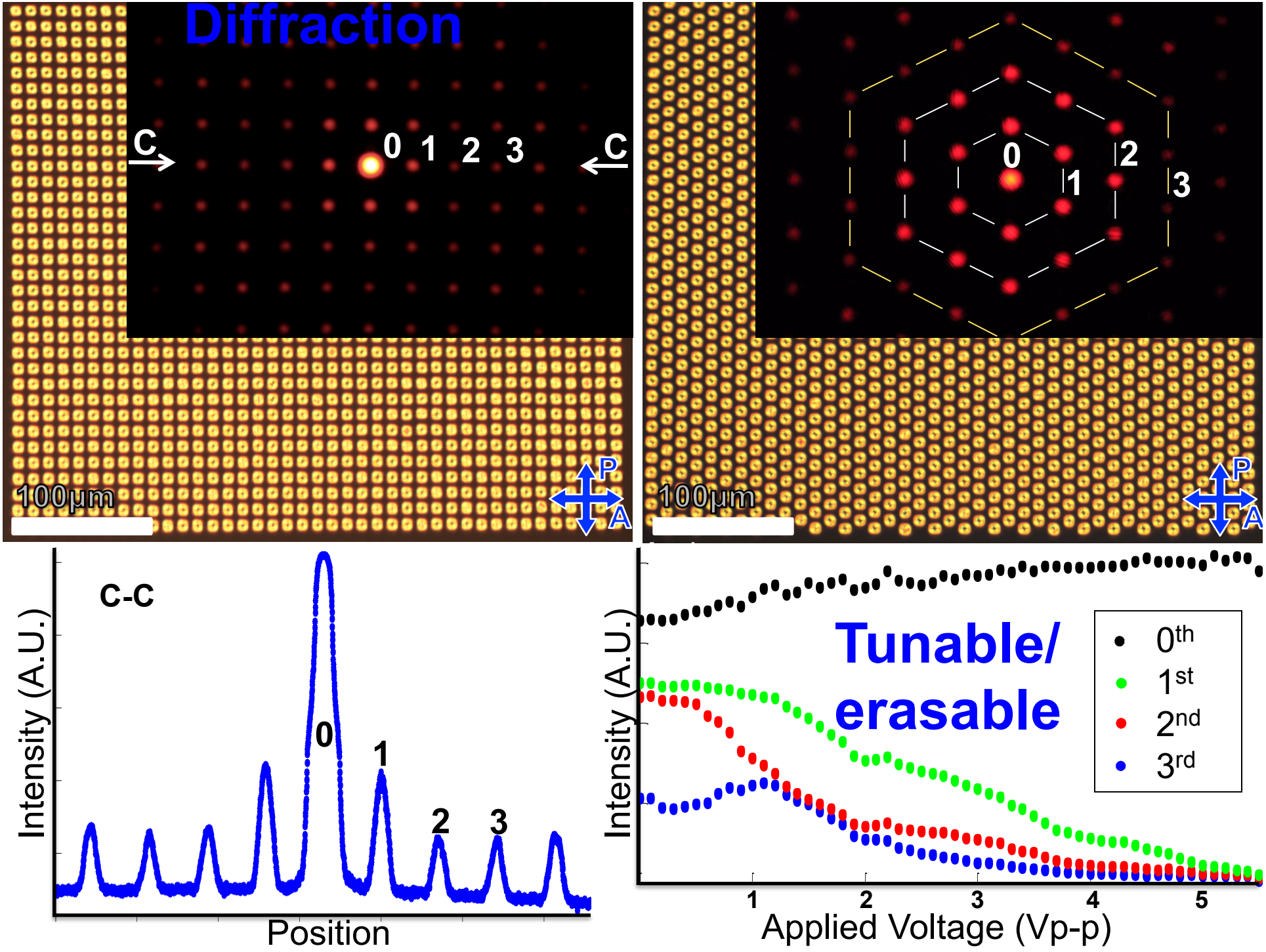
Periodic lattices & quasicrystal-like structures

Honeycomb lattice of torons

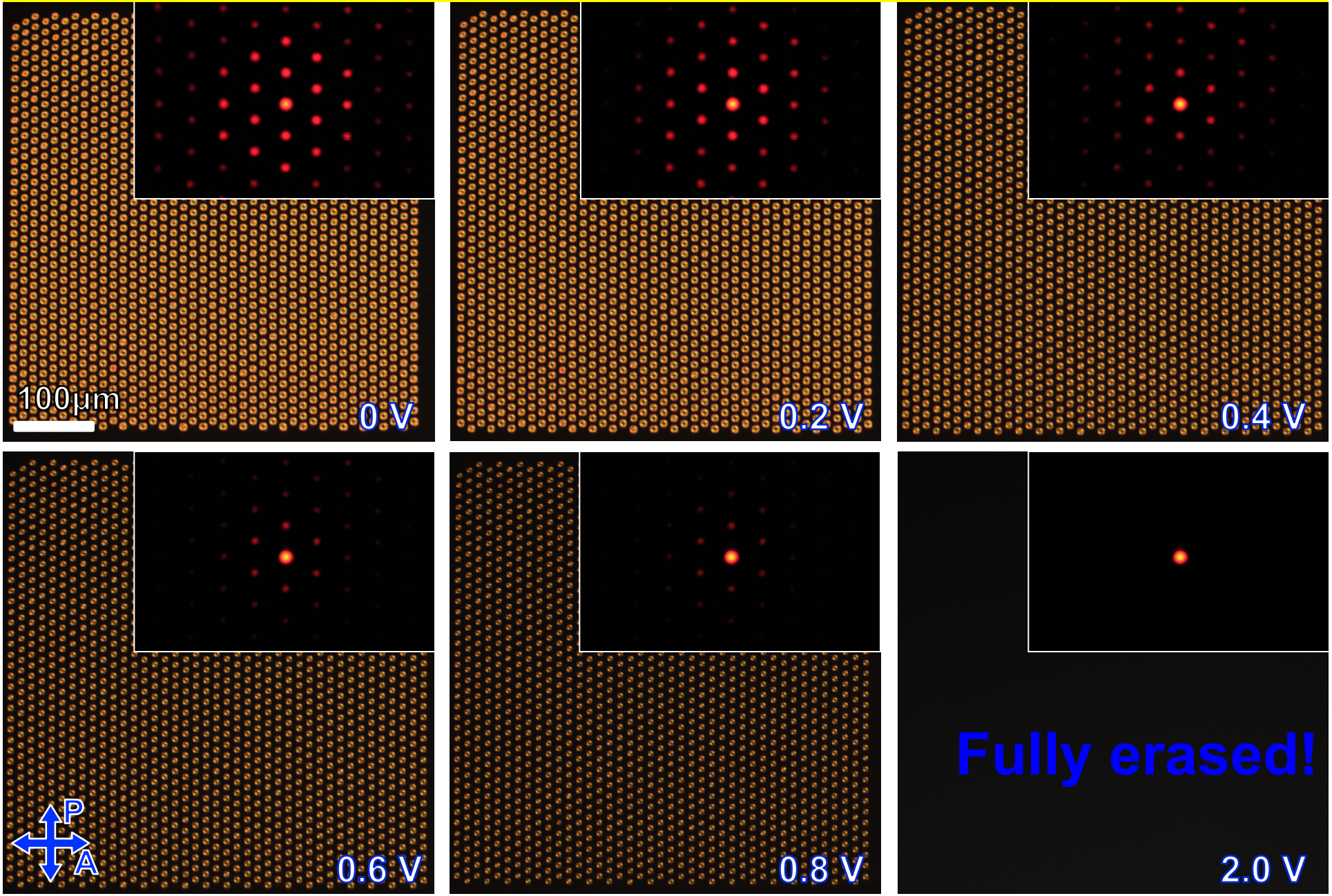
Quasicrystal-like structure



Nature **454**, 501-504;
Ackerman, Qi, Smalyukh, Phys Rev E (2012)

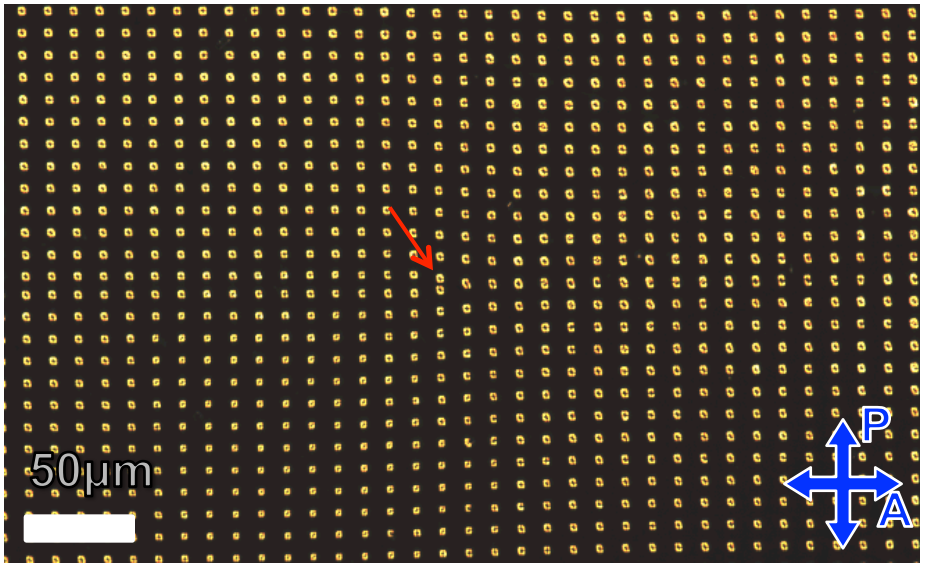

Diffraction
C
C
0 1 2 3
100μm
P
A
0
1
2
3
100μm
P
A
C-C
Intensity (A.U.)
0
1
2
3
Position
Tunable/
erasable
0th
1st
2nd
3rd
Intensity (A.U.)
1
2
3
4
5
Applied Voltage (Vp-p)

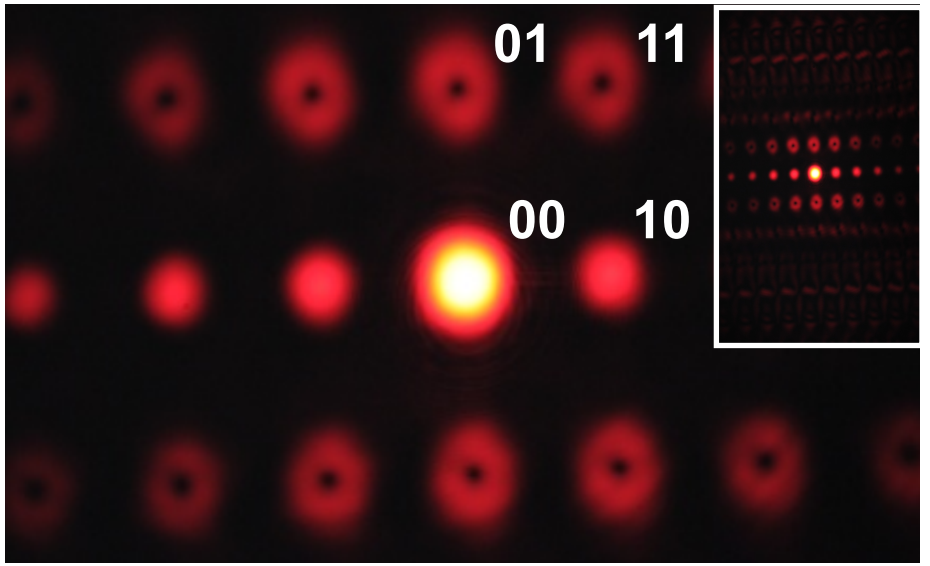
Example of tuning/erasing toron gratings

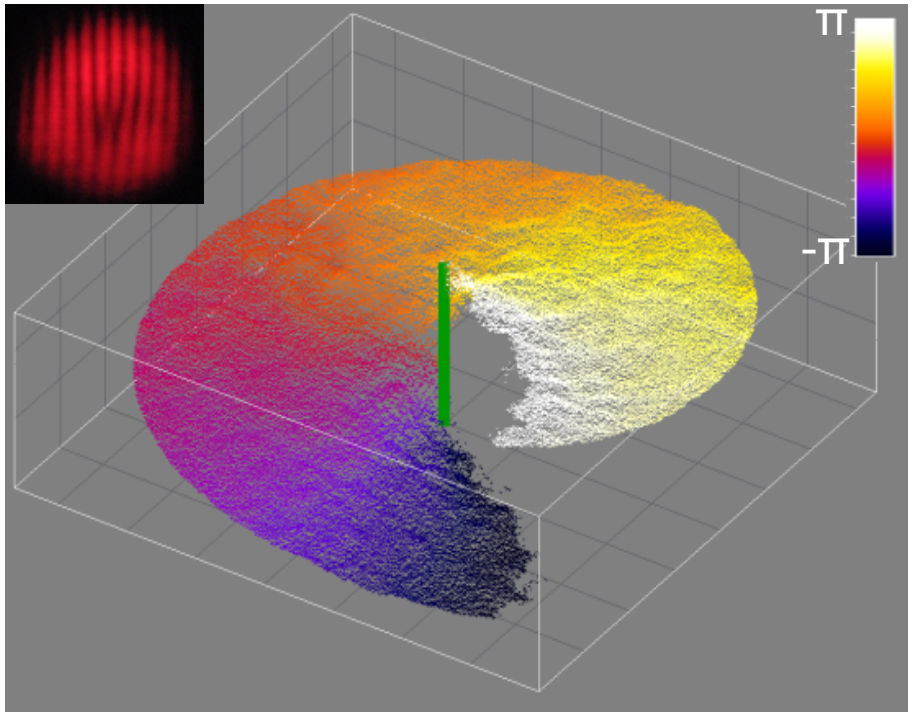


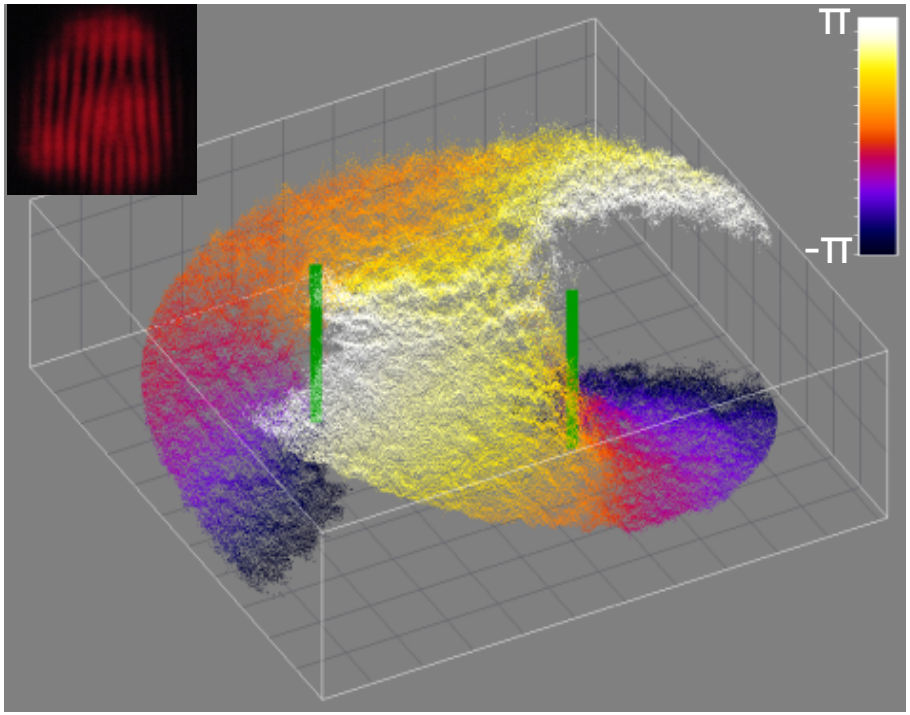
Ackerman, Qi, & Smalyukh, Phys Rev E (2012)

Phase singularities generated by toron arrays

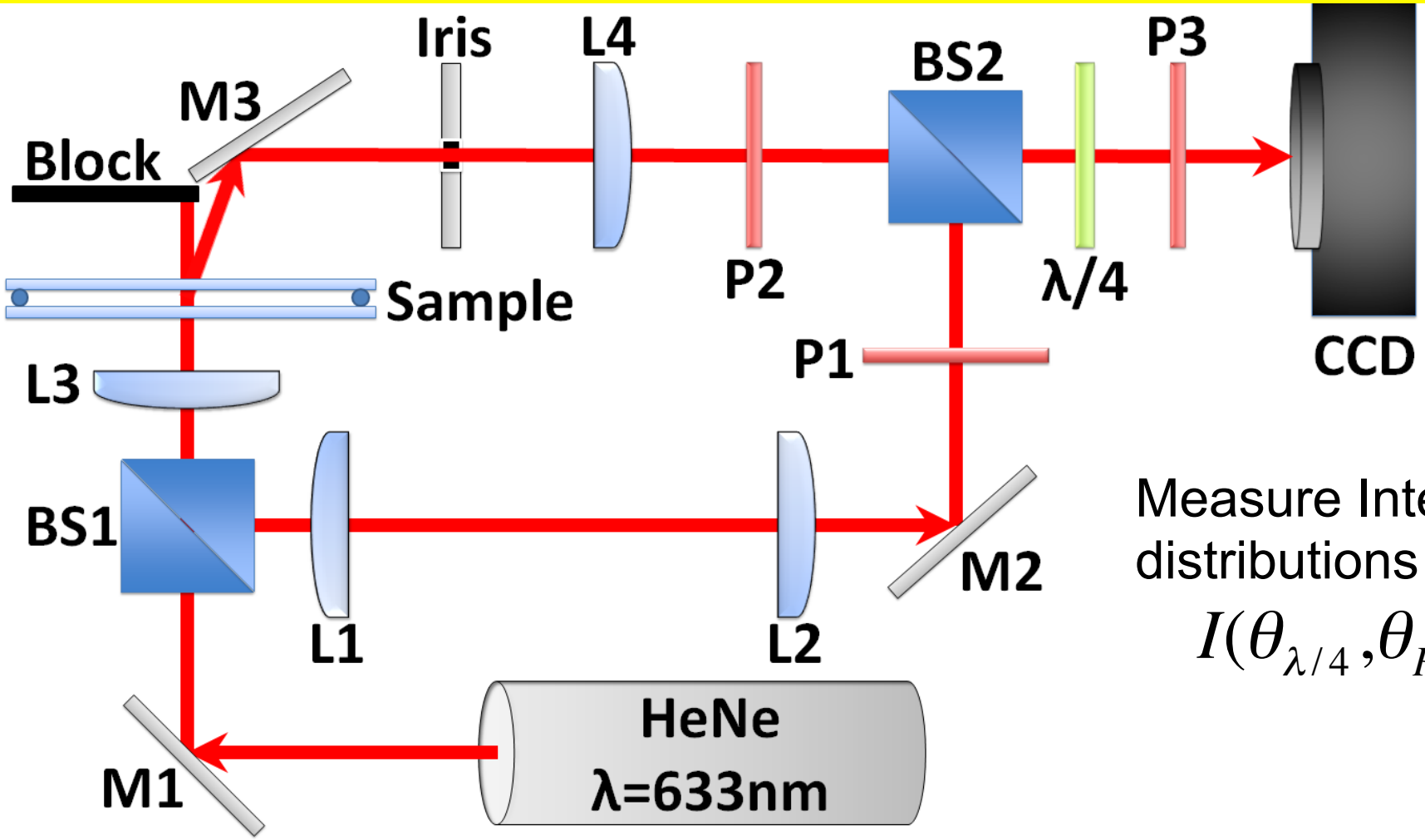








Stokes Polarimetry for Phase Mapping



Measure Intensity distributions

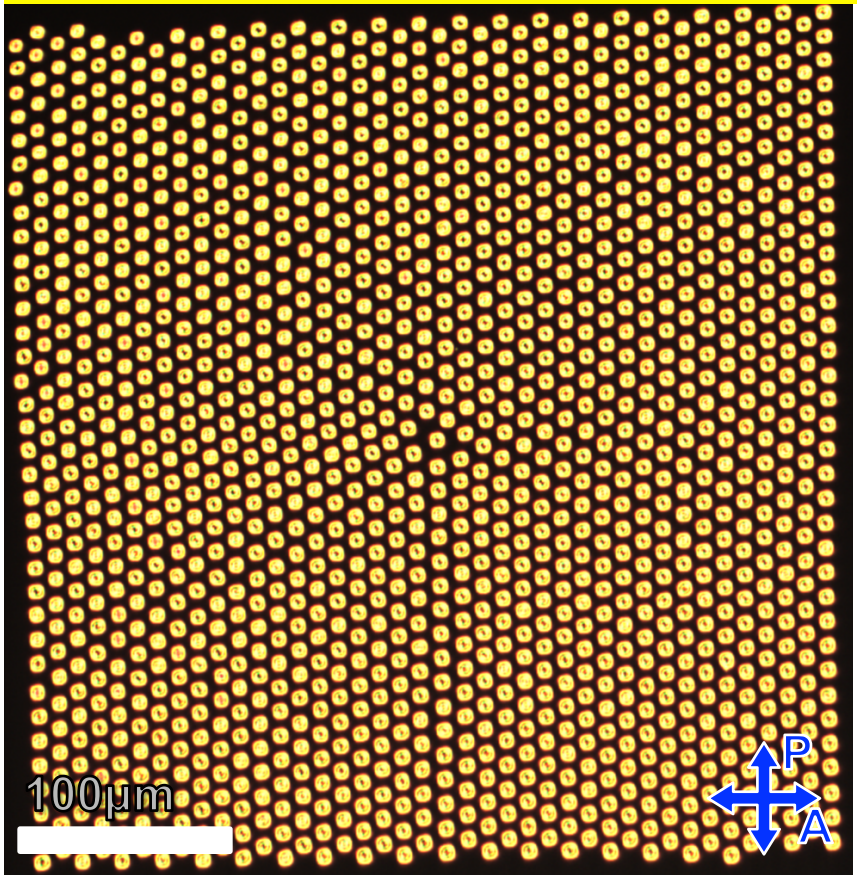
$$I(\theta_{\lambda/4},\theta_P)$$

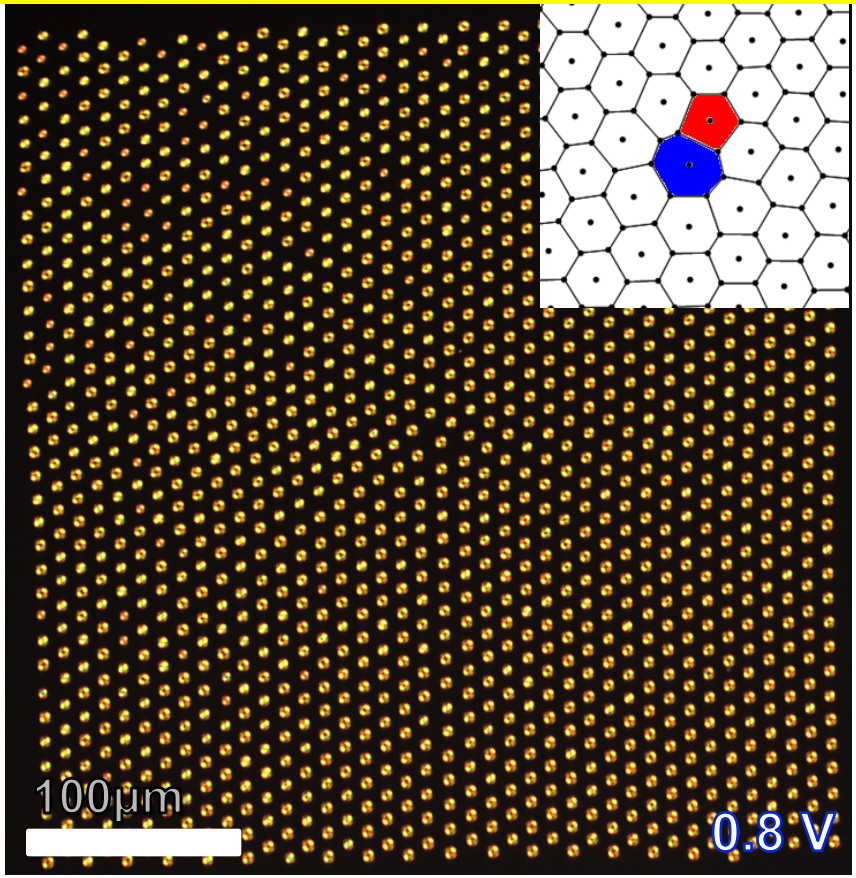
$$S_2 = I(\pi/4,\pi/4) - I(3\pi/4,3\pi/4) \qquad S_3 = I(\pi/4,0) - I(3\pi/4,0)$$

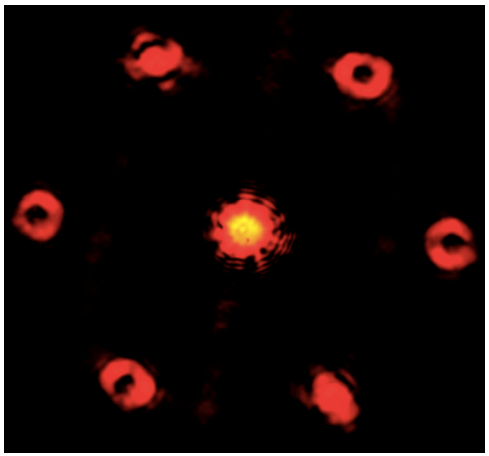
Phase profile $\delta(x,y) = \arctan[S_3(x,y)/S_2(x,y)]$

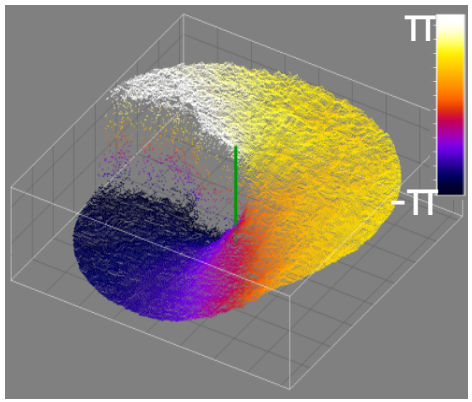
Denisenko *et al.*, *Opt. Lett.* **33**, 89–91 (2008).

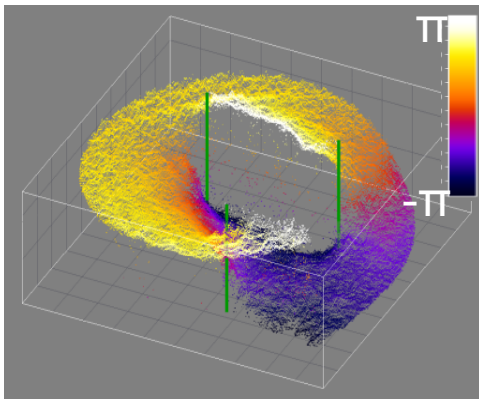
5-7 defects in toron arrays & phase singularities

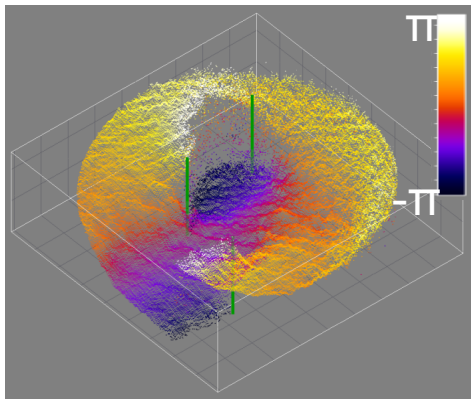




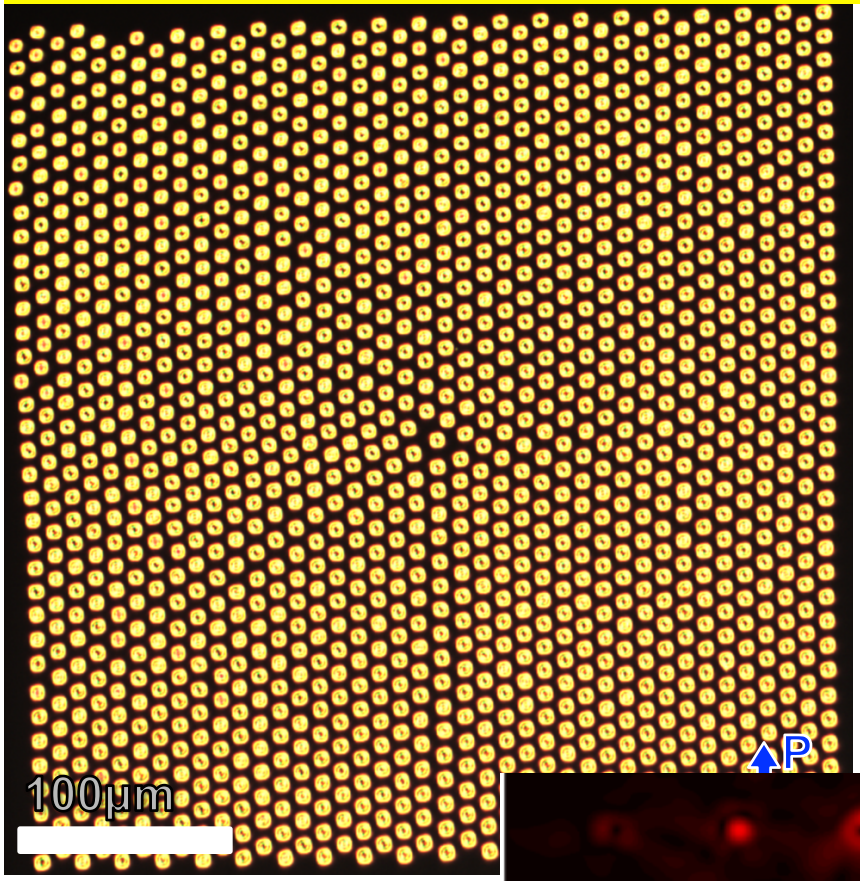


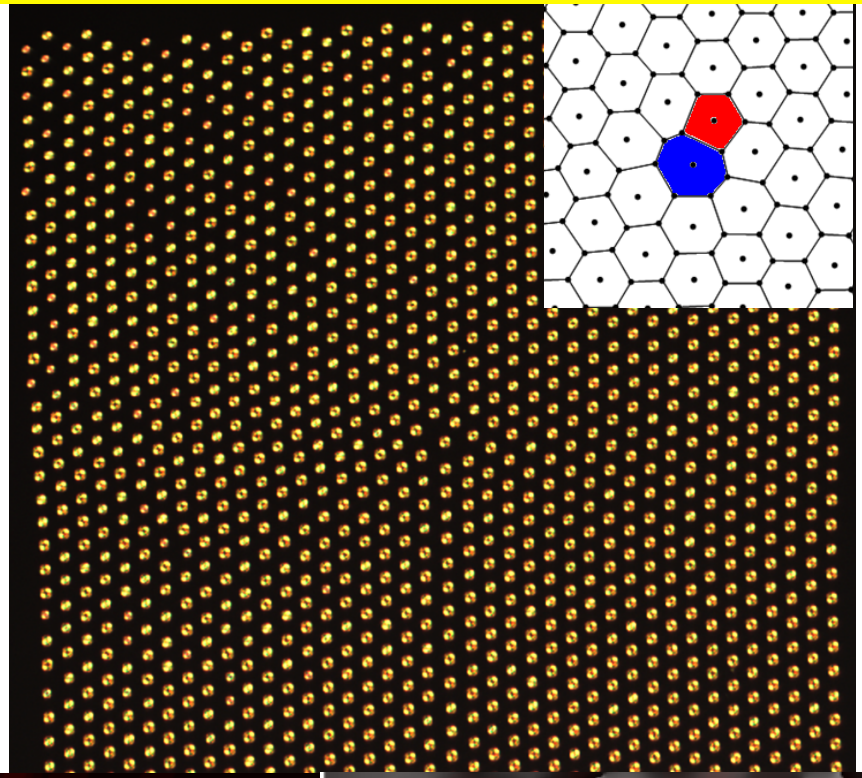


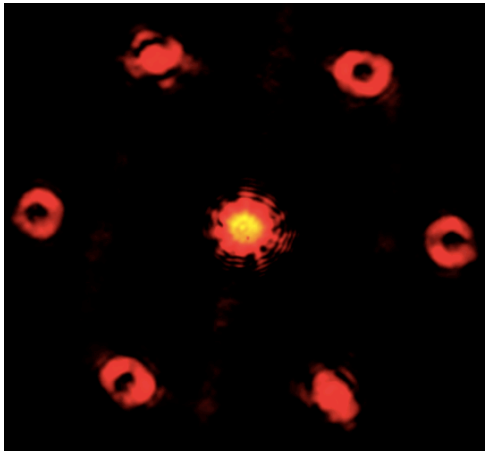


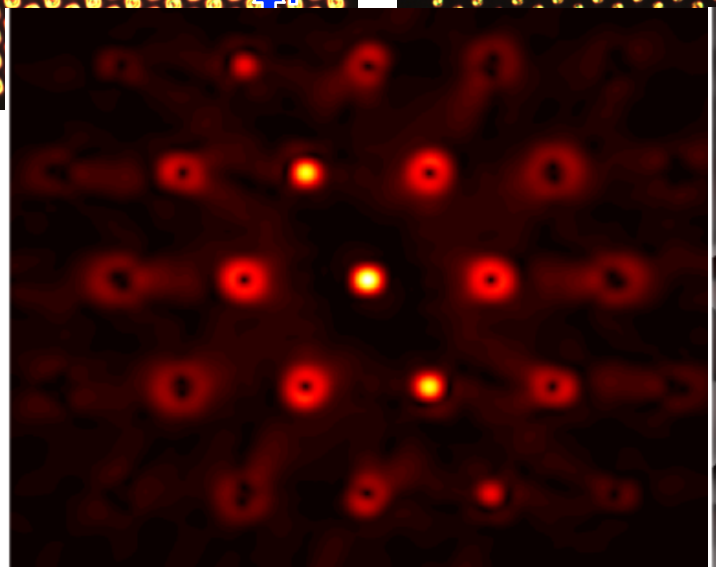


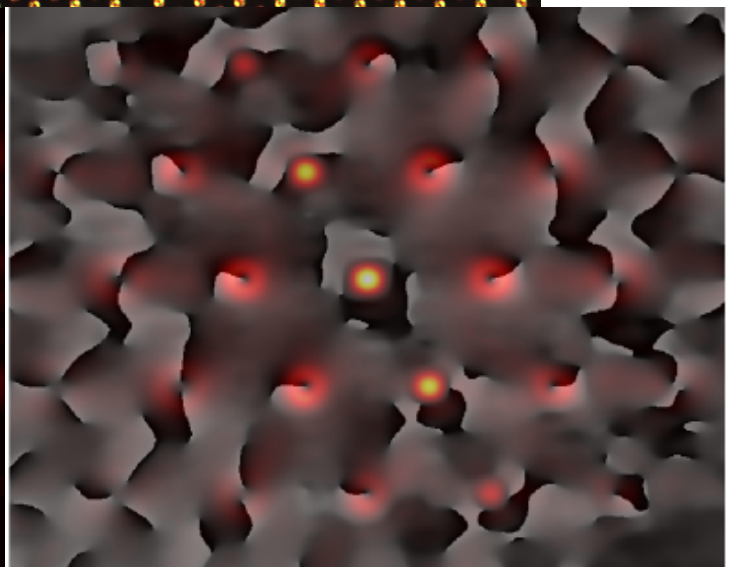
5-7 defects in toron arrays & phase singularities





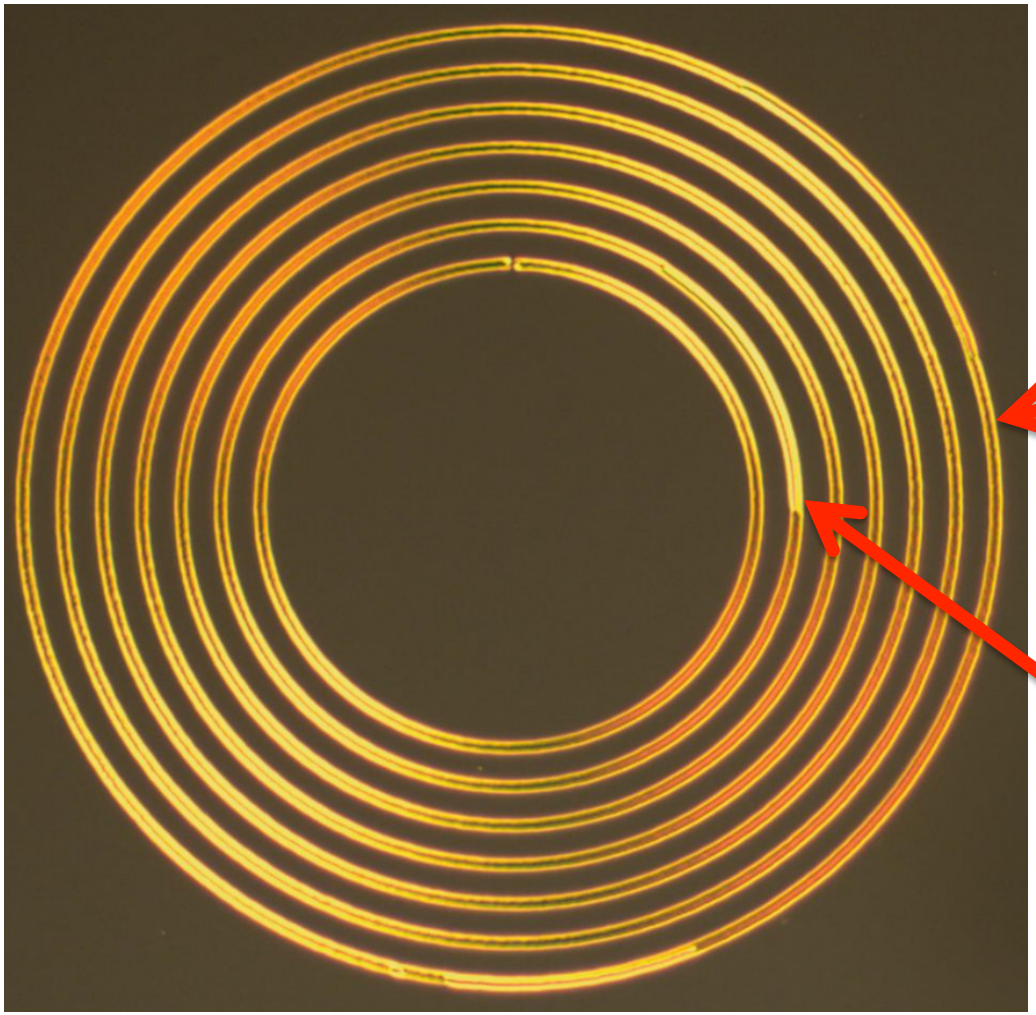




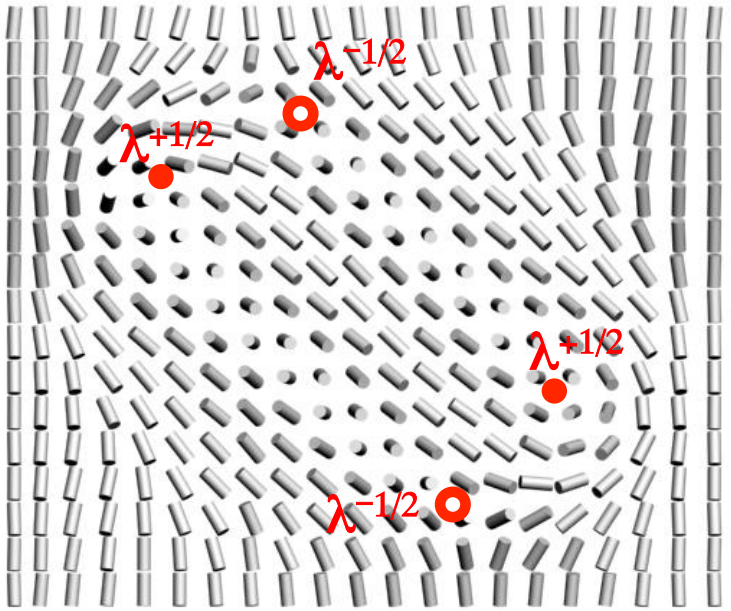


Optically generated stable cholesteric fingers

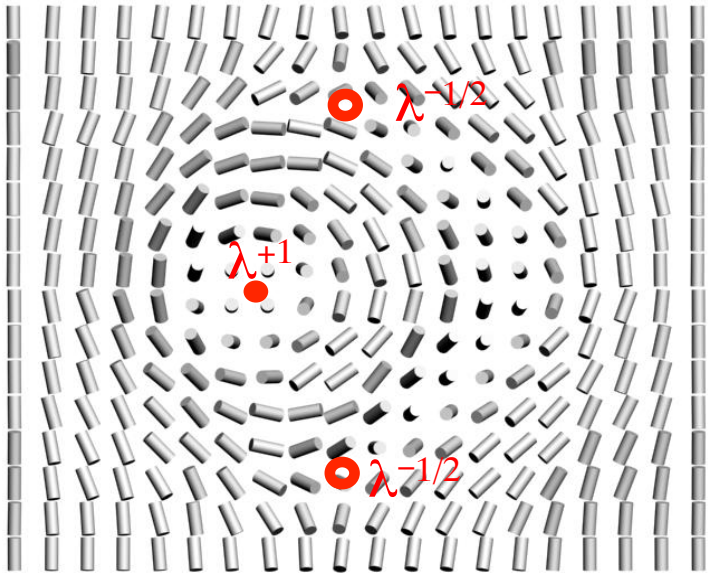
By continuously dragging the IR beam



CF1

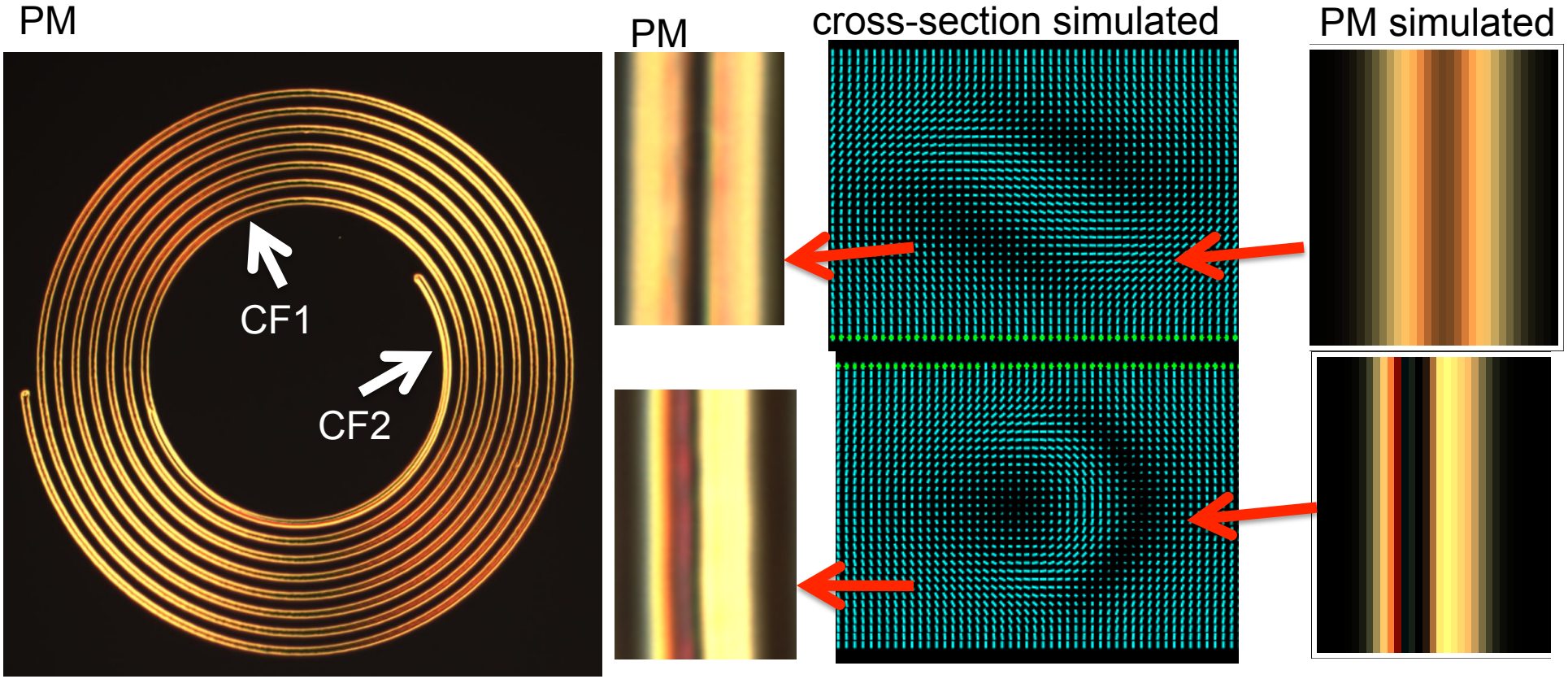


CF2

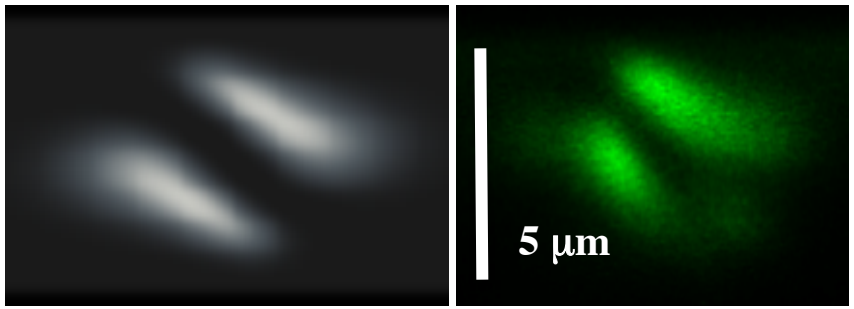


→Strength conserved;
→Minimizes twist elastic energy in the expense of bend & splay;

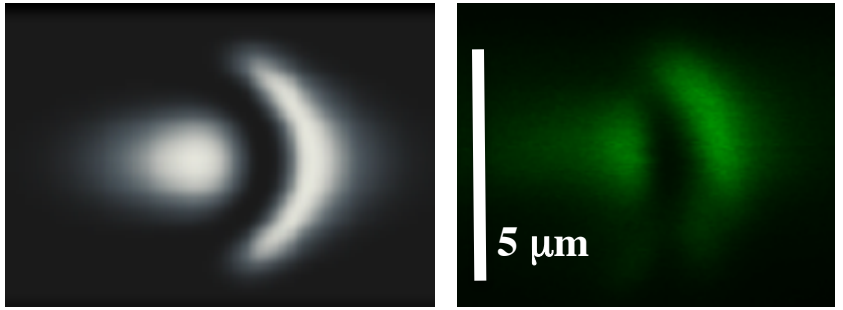
Cholesteric fingers: simulations vs. experiments



3PEF-PM image experimental/simulated

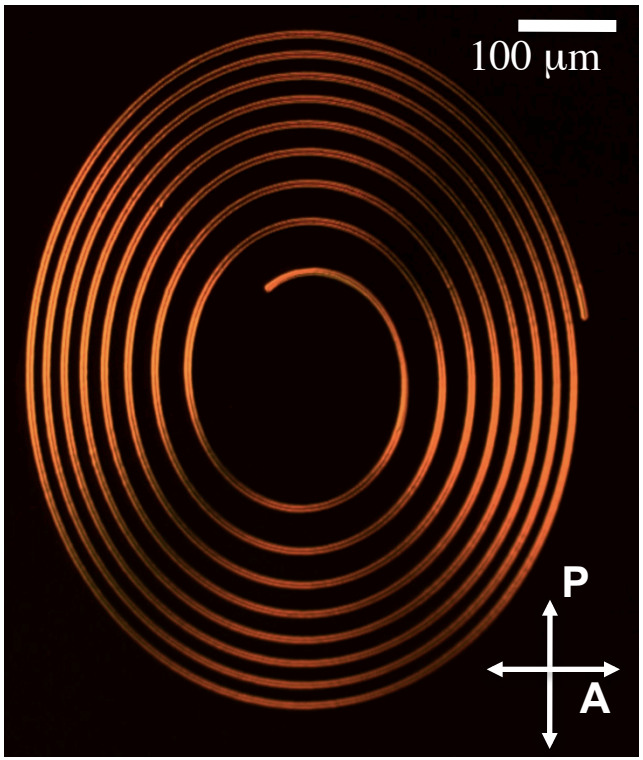


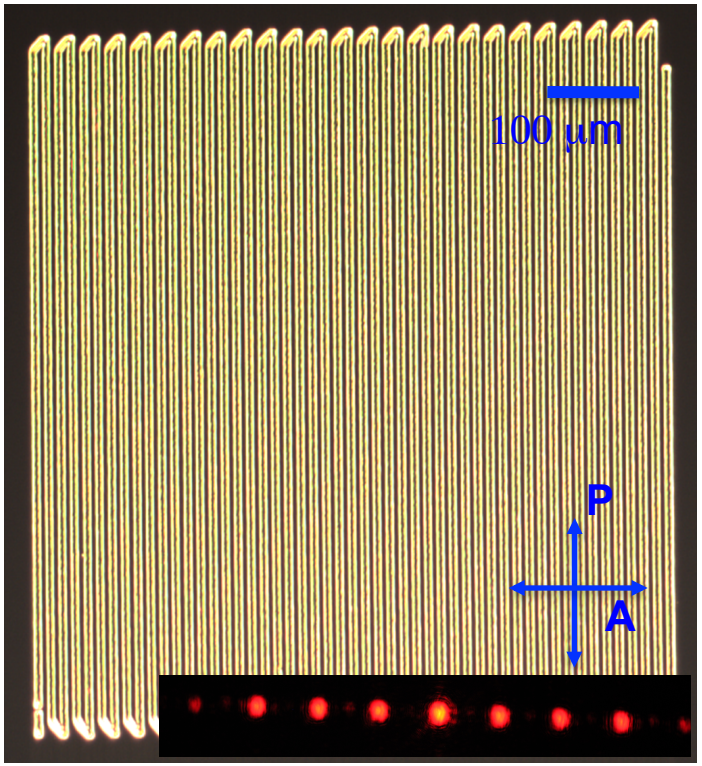
3PEF-PM image experimental/simulated

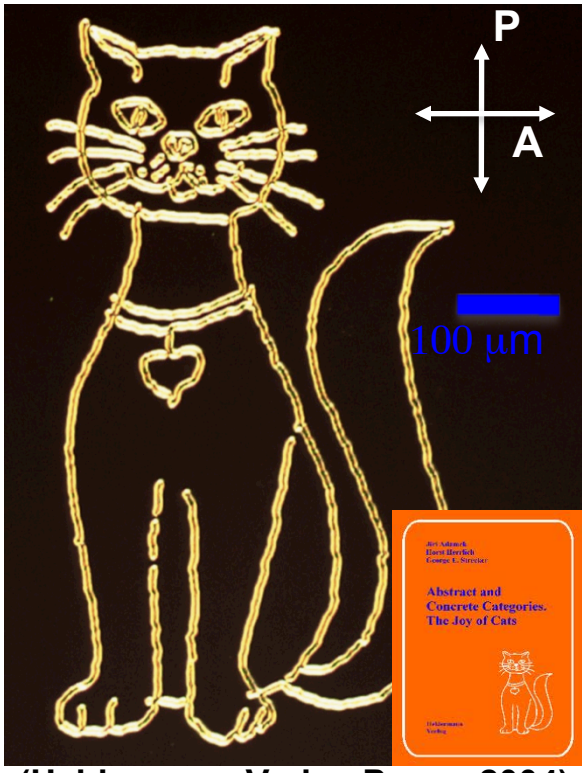


Ackerman, Qi, Lin, Twombly, Laviada, Lansac, Smalyukh, *Scientific Reports* **2**, 414 (2012)

Optical drawing of twist-escaped disclinations

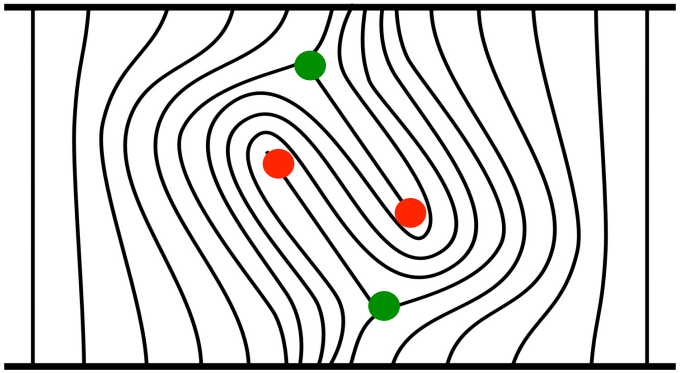




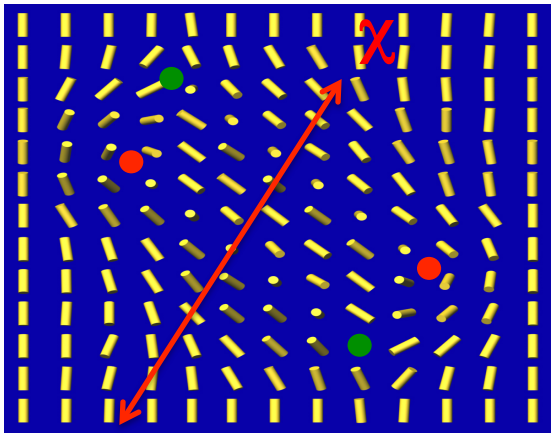


Adamek, Herrlich, Strecker,*Abstract and Concrete Categories-The Joy of Cats*. (Heldermann Verlag Press, 2004).

Unstable cluster of 4 nematic half-integer disclinations

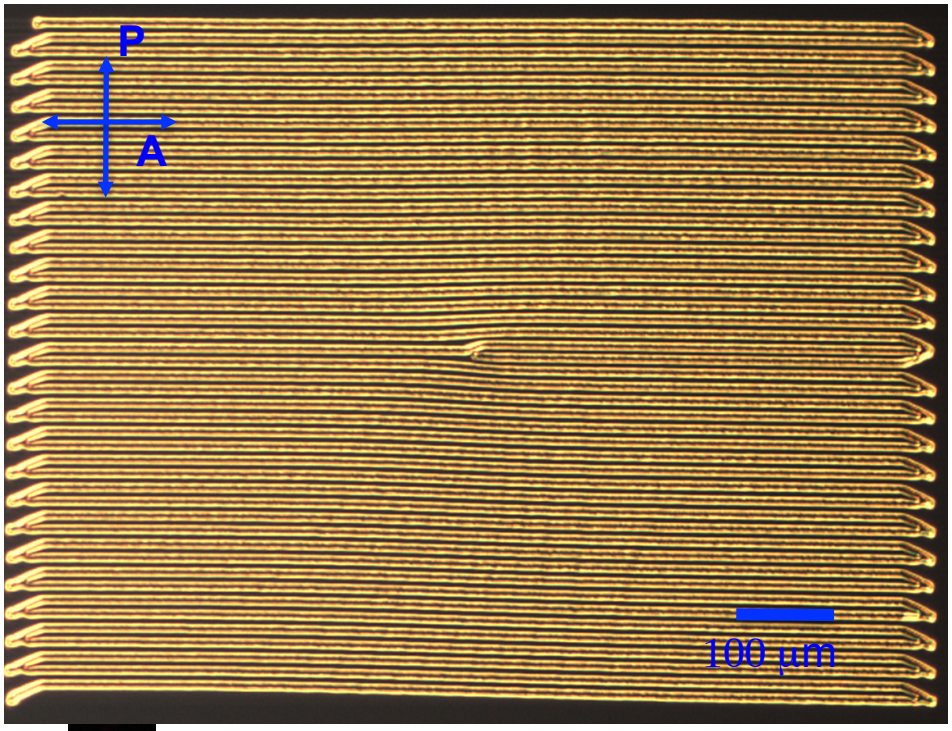


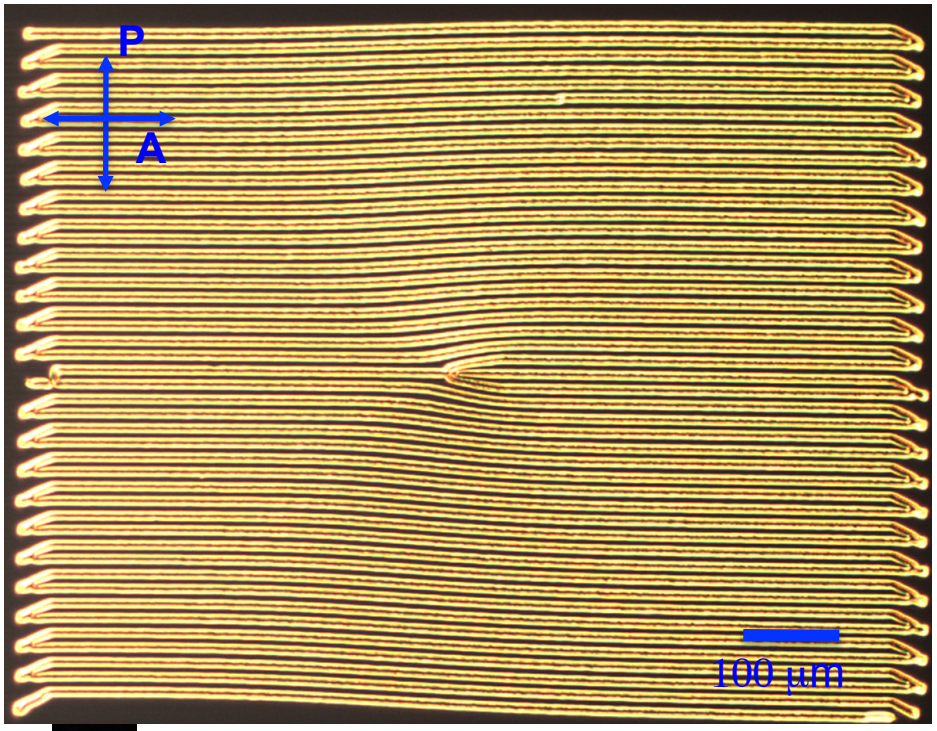
Twist-stabilized disclinations

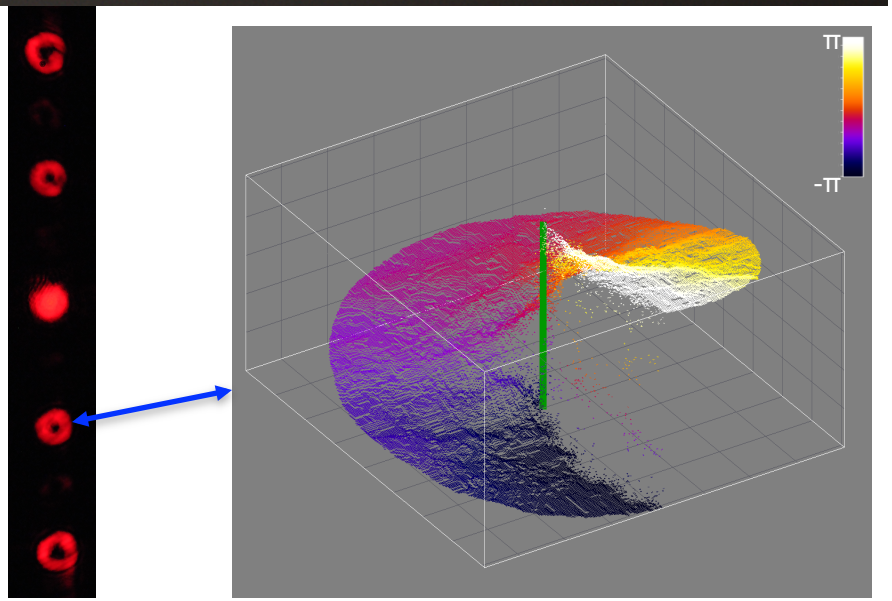


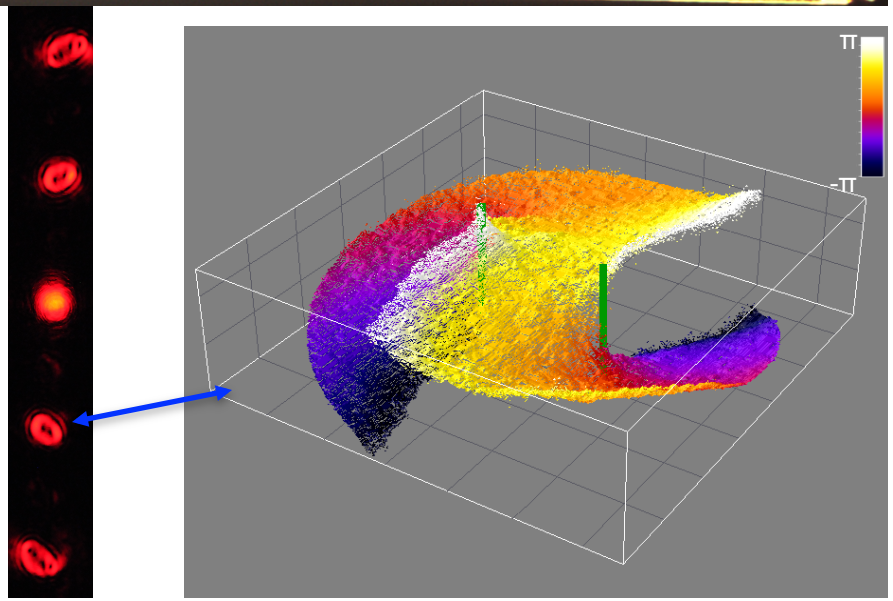
Can selectively draw structures from CF1 or CF2

Optical vortices vs. Burgers vector of finger gratings



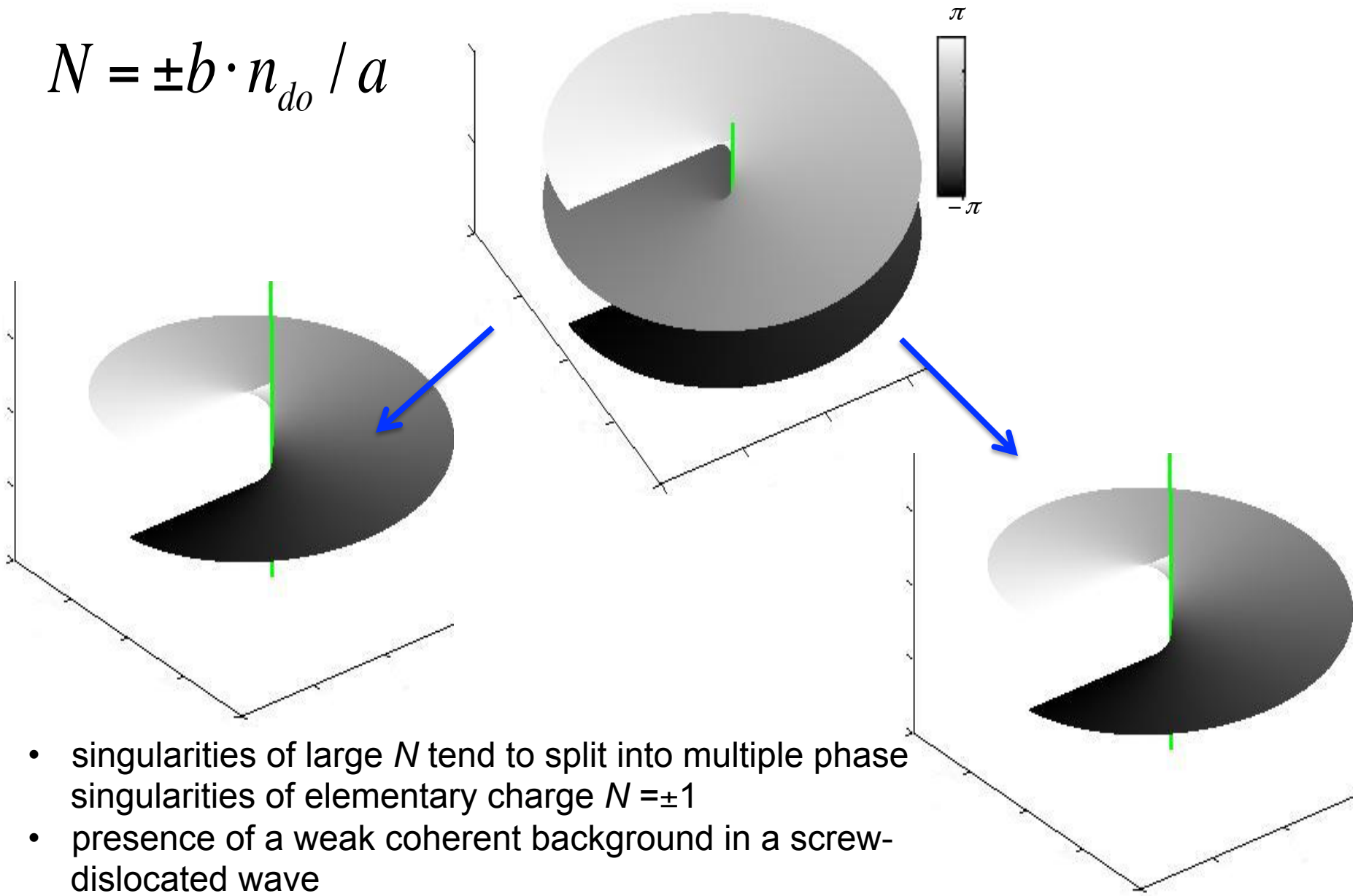






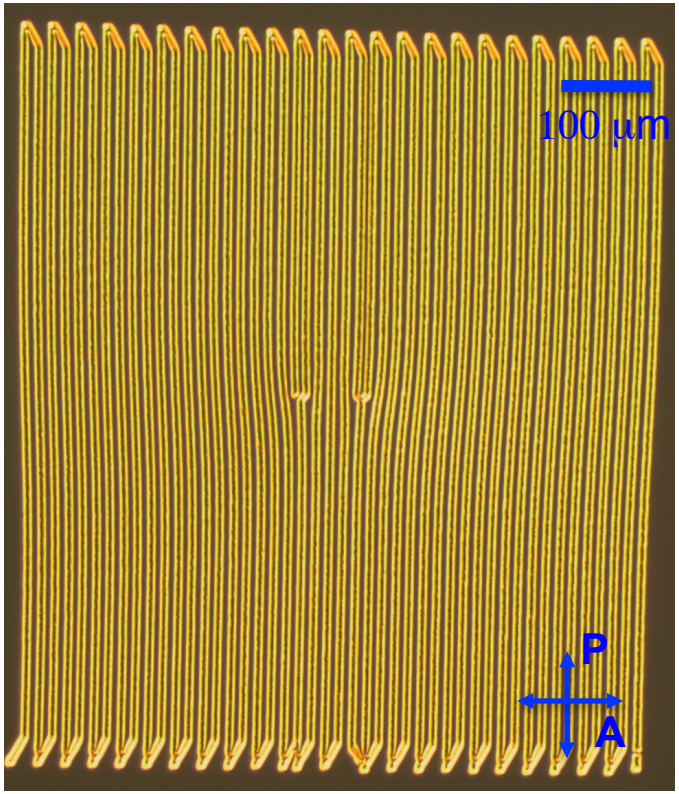
Splitting of high-charge phase singularities

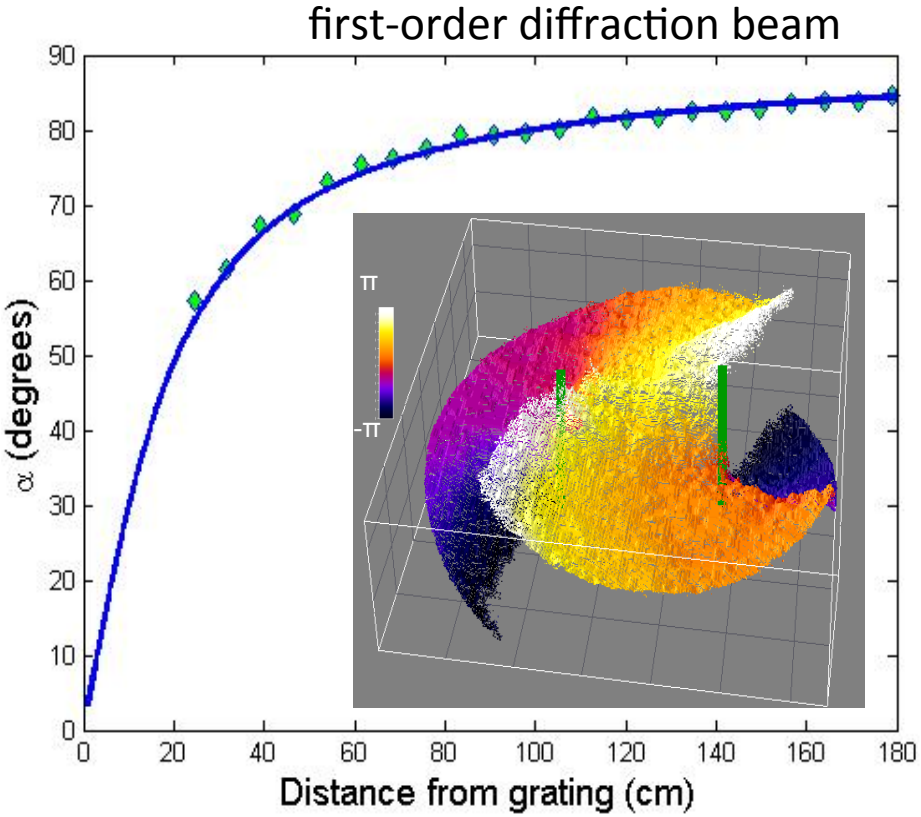
$$N = \pm b \cdot n_{do} / a$$

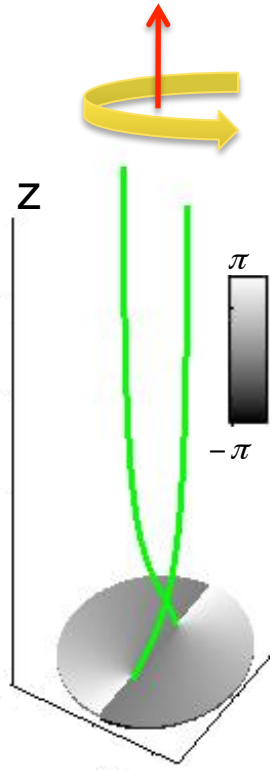


- singularities of large *N* tend to split into multiple phase singularities of elementary charge *N* =±1
- presence of a weak coherent background in a screw-dislocated wave

Twisting vortices induced by defects in a finger grating







Gouy phase shift angle

$$\alpha = \arctan(d_c / d_R) + \alpha_{offset}$$

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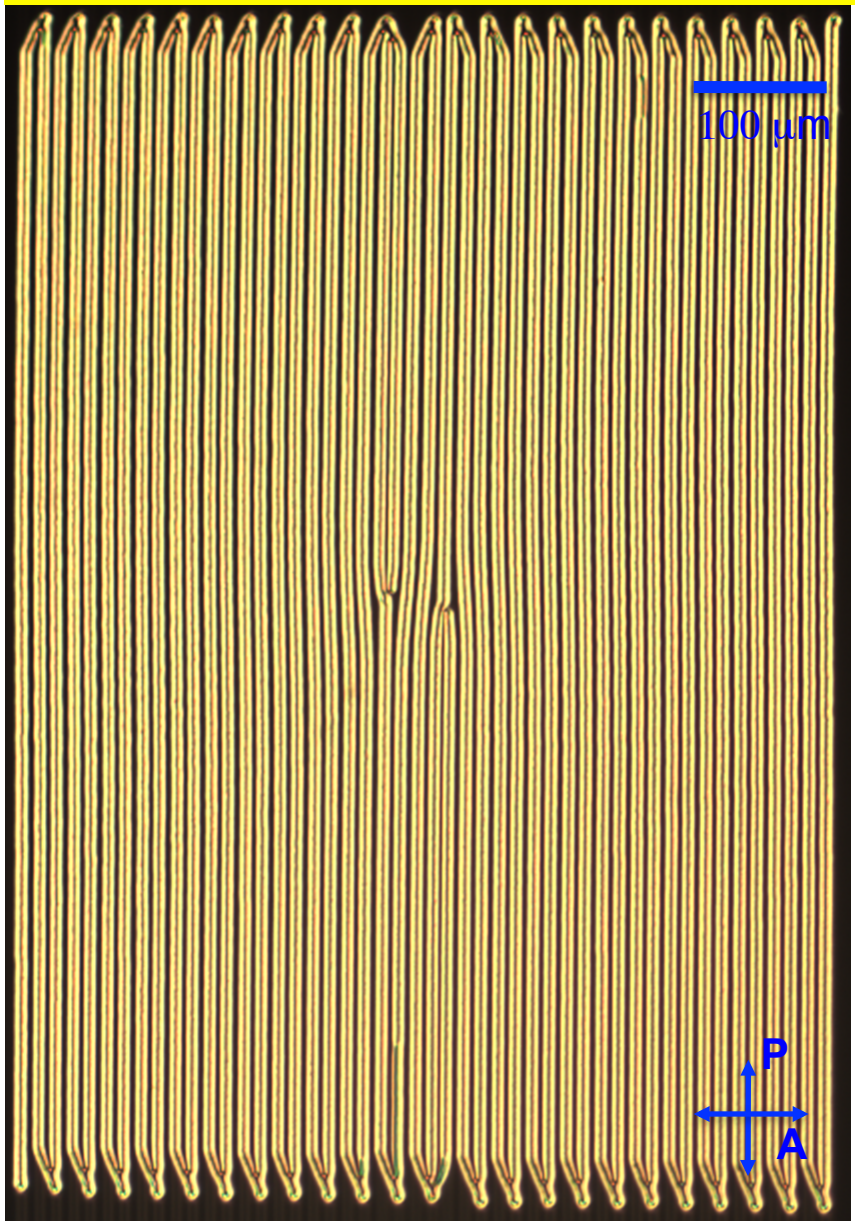
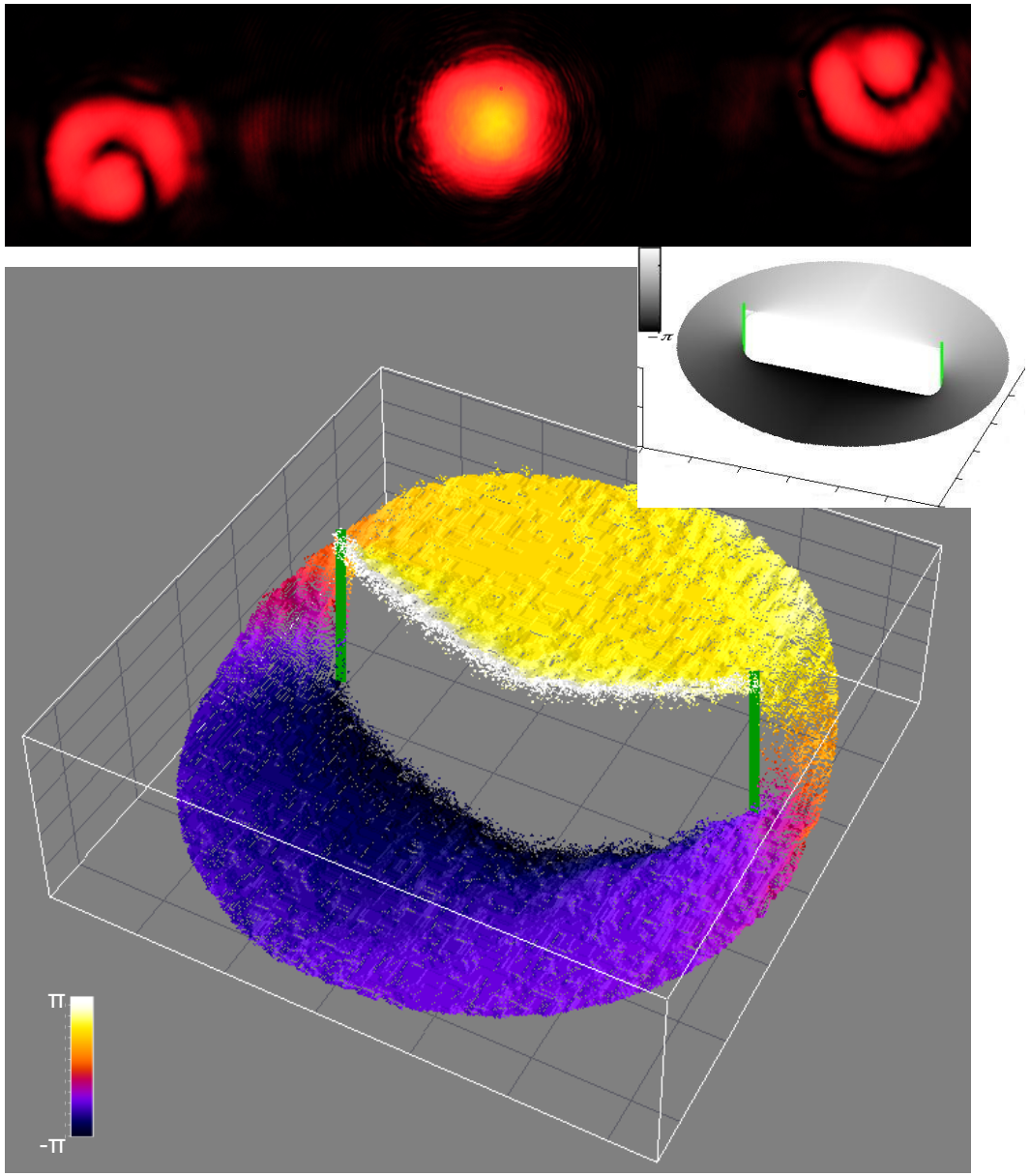
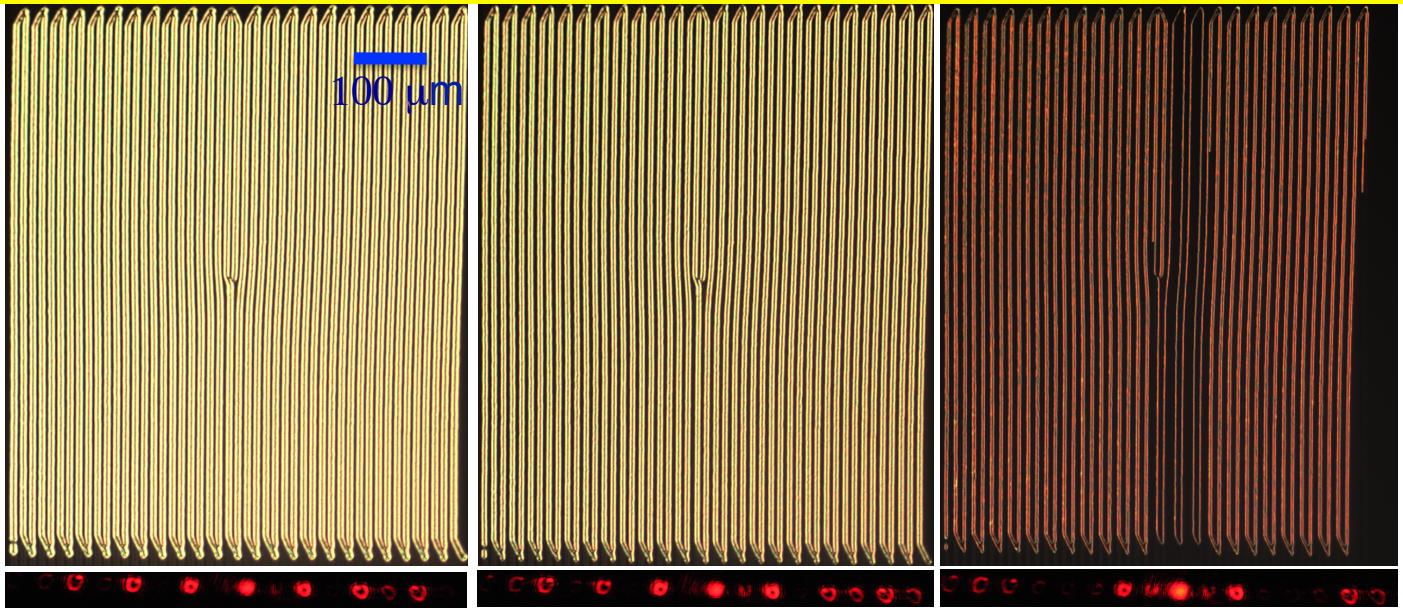
Inter-linked edge-screw dislocations

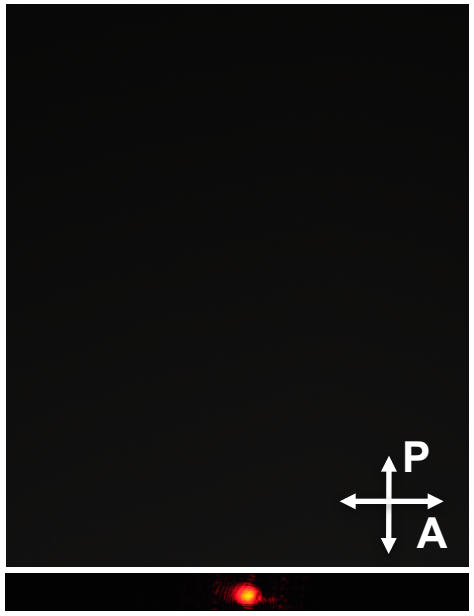


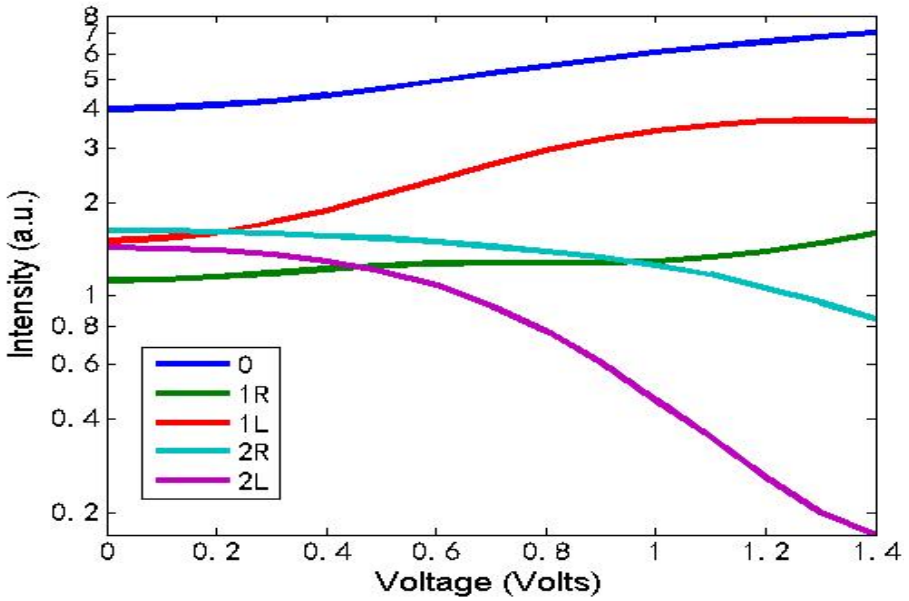


Ackerman, Qi, Lin, Twombly, Laviada, Lansac, Smalyukh, *Scientific Reports* **2**, 414 (2012)

Voltages control of fingers gratings and the corresponding diffraction patterns

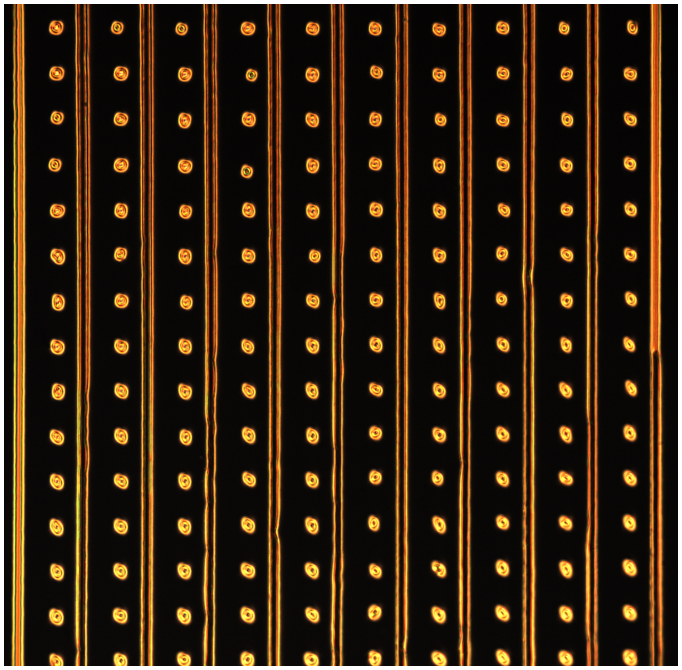




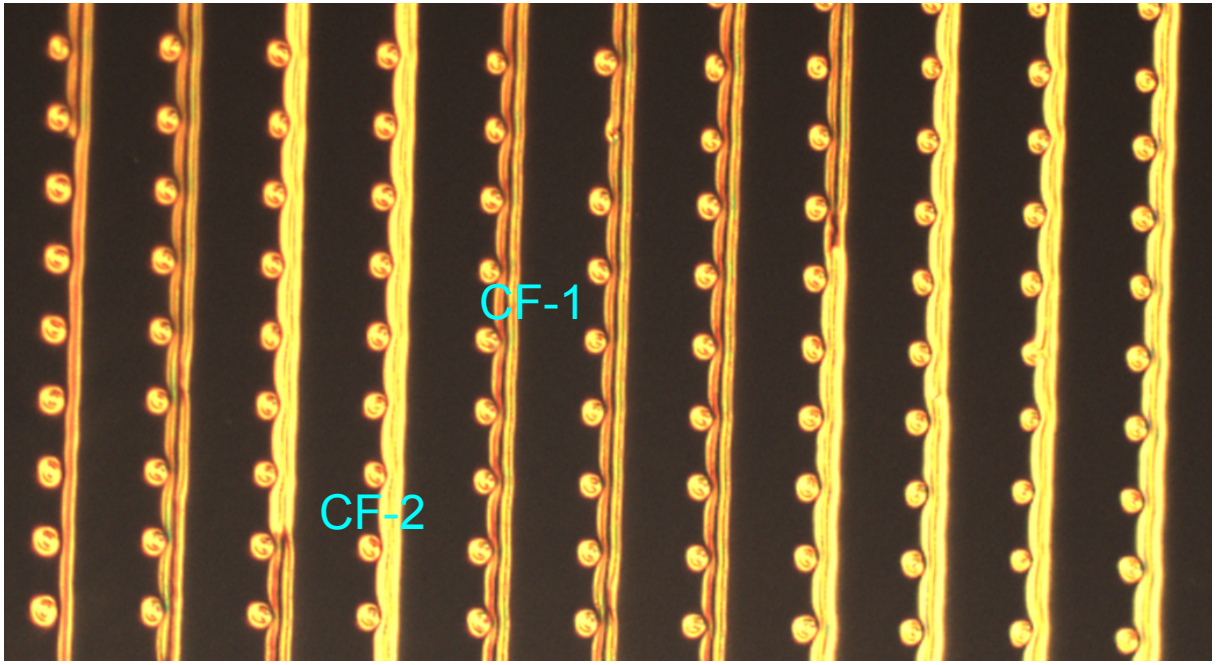


Toron-finger elasticity-mediated interactions

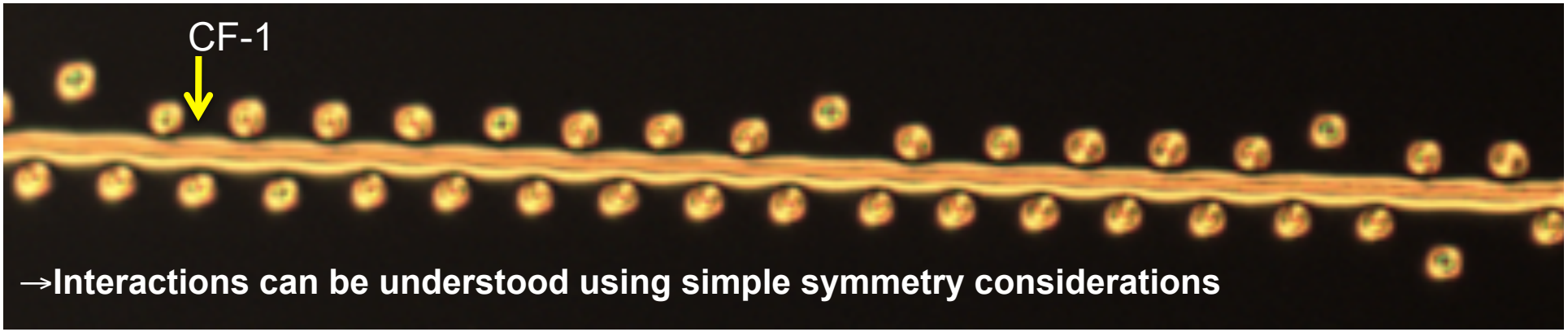
→Repulsive/weak Interactions



→Attractive Interactions

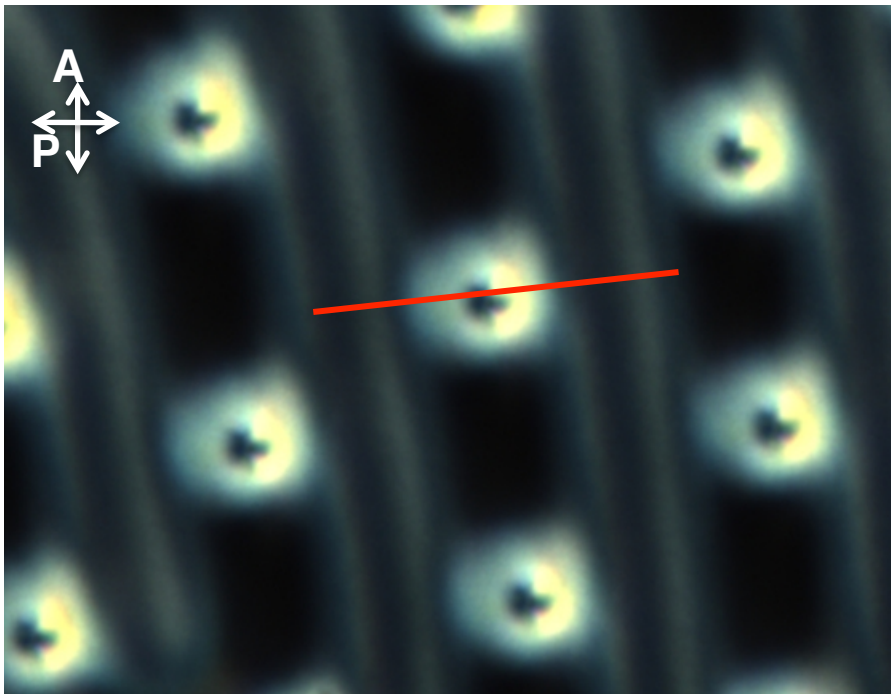


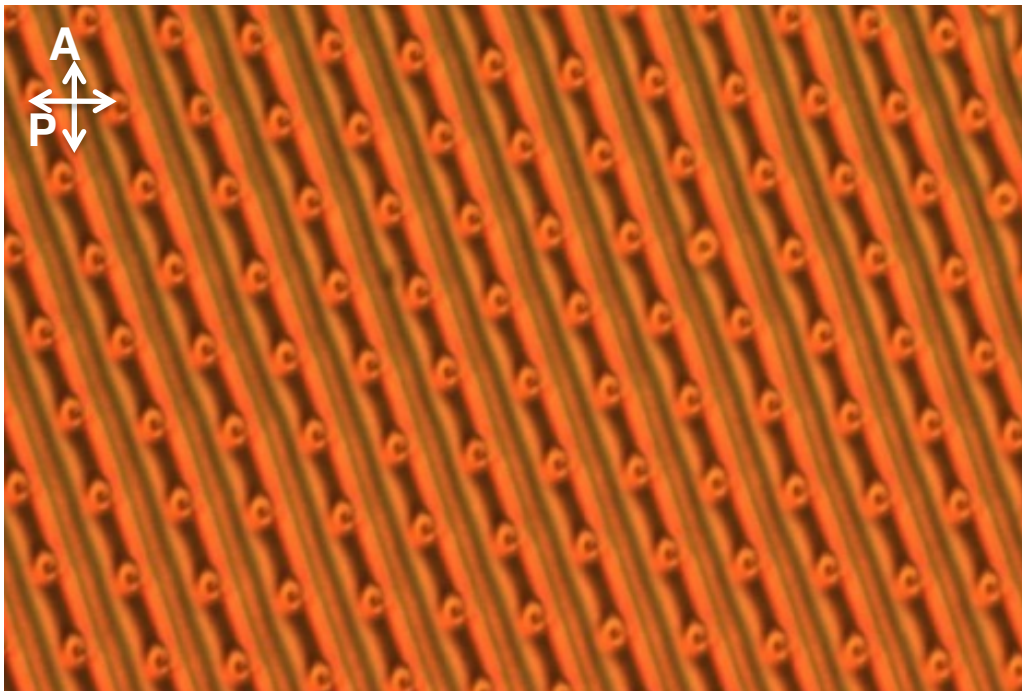
→Strong pinning vs. attractive toron-finger interactions and self-assembly



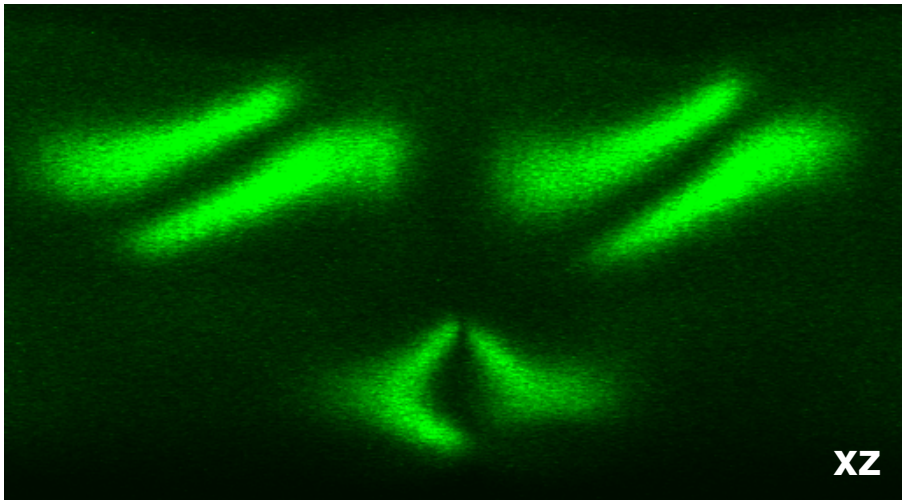
Ackerman, Qi, Lin, Twombly, Laviada, Lansac, Smalyukh, *Scientific Reports* **2**, 414 (2012)

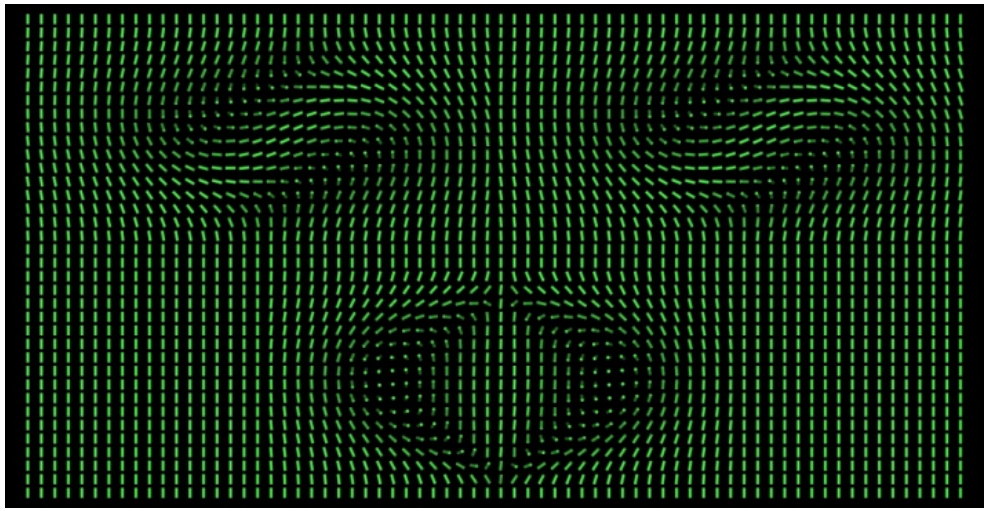
3D templated self-assembly of torons & fingers





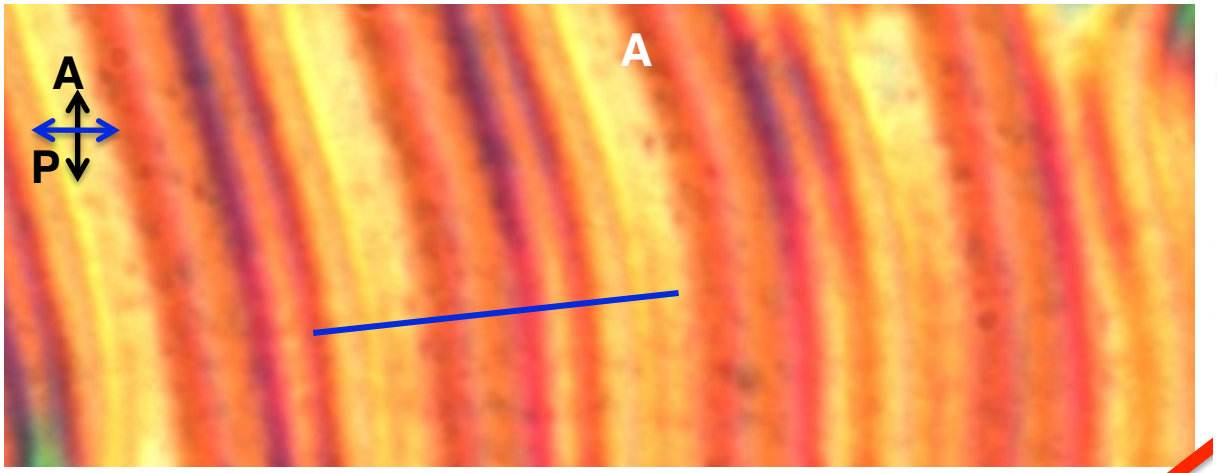
Reconstructed director field

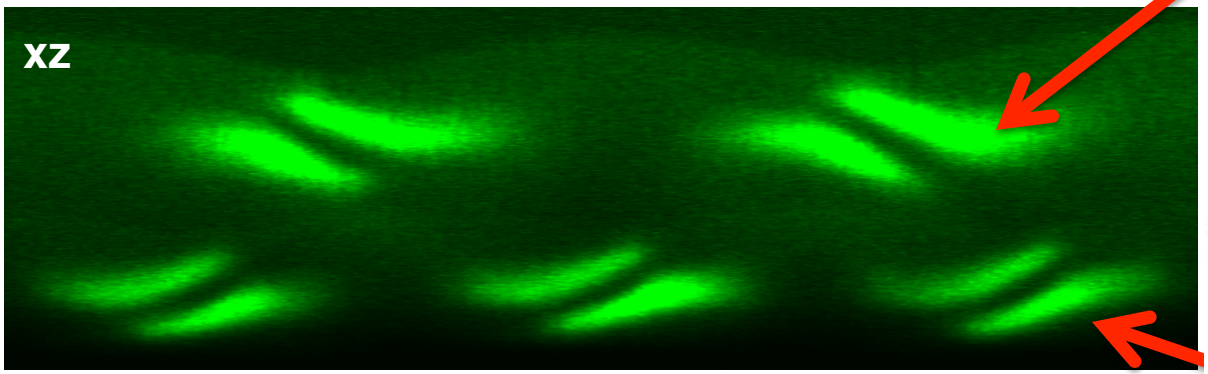


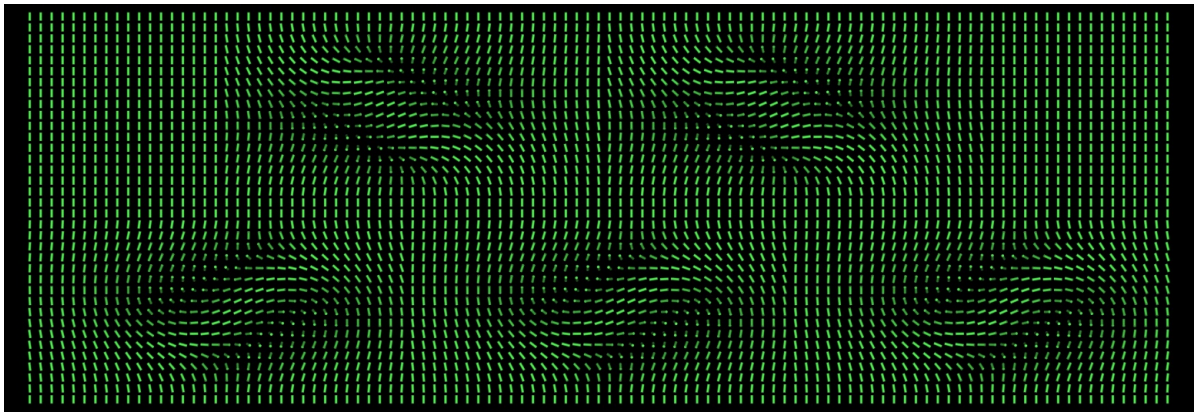


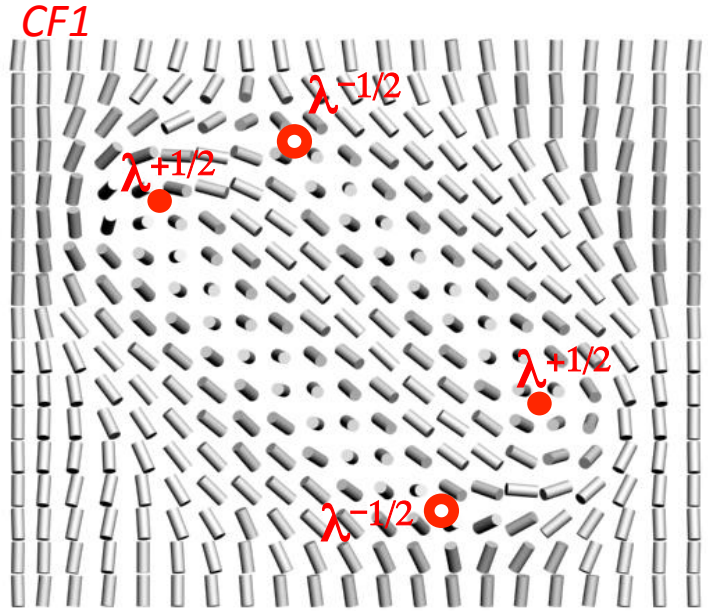
Evans, Ackerman, Broer, van de Lagemaat, Smalyukh, *Phys Rev E* (2012)

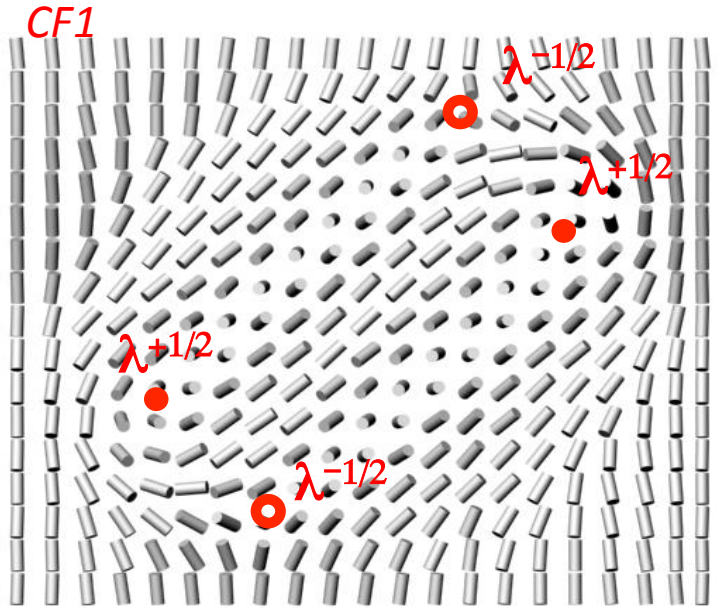
Layer-by-layer self-assembly of finger arrays



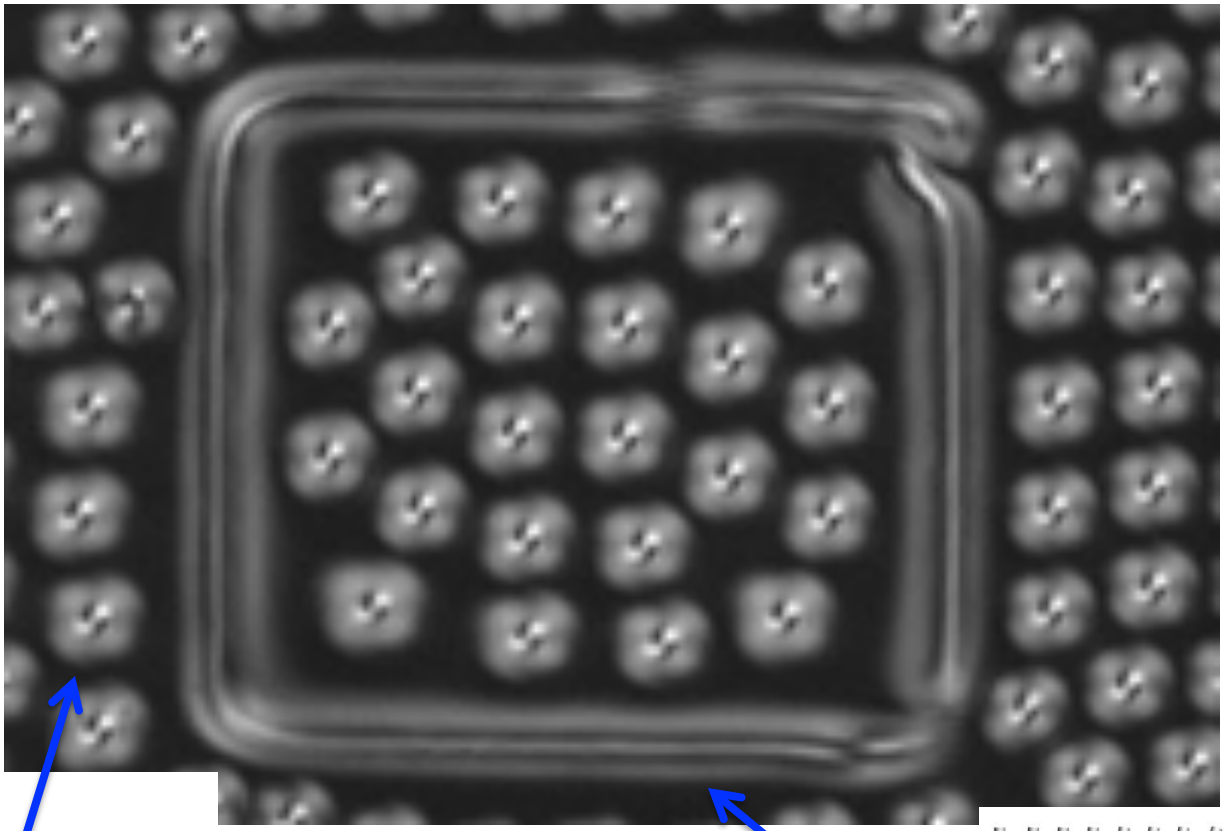


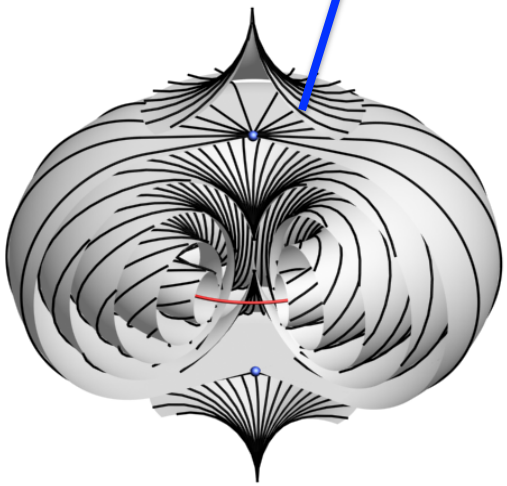




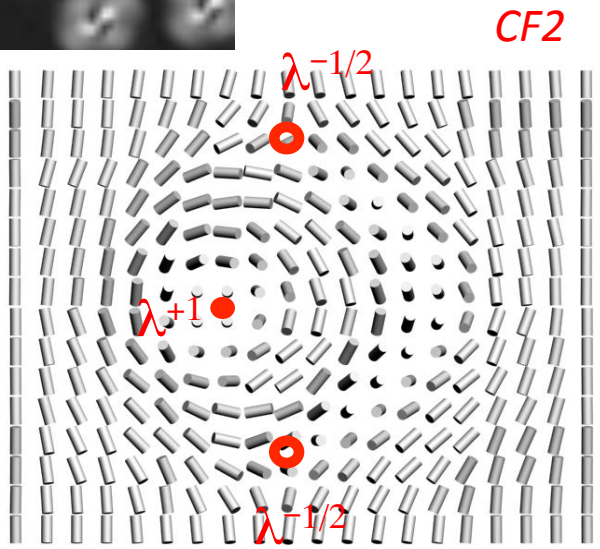


Topological Dance





- Torons & fingers CF2 dance when exposed to scanned linearly polarized laser light;
- "Baby-Skyrmions" have never been spotted dancing!



I.I. Smalyukh et al, *Opt. Express* **20**, 6870-6880 (2012).

Conclusions & outlook

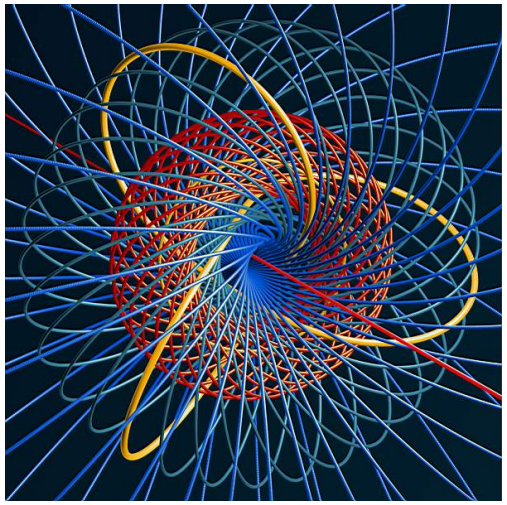
→ Topological defects in LCs controlled by beams with optical vortices;

→ Optical vortices controlled by defects in hierarchical ordered structures of defects;

→Crystals, Quasicrystals & other 3D assemblies of twist-bound defects;

→Elasticity-mediated self-assembly of torons, scyrmions, & fingers, merons, etc.;

→Quasicrystals of Seifert fibrations next time?



Source: math.cnrs.fr

Thank you !!!