Topological Colloids & Liquid Crystal Skyrmions

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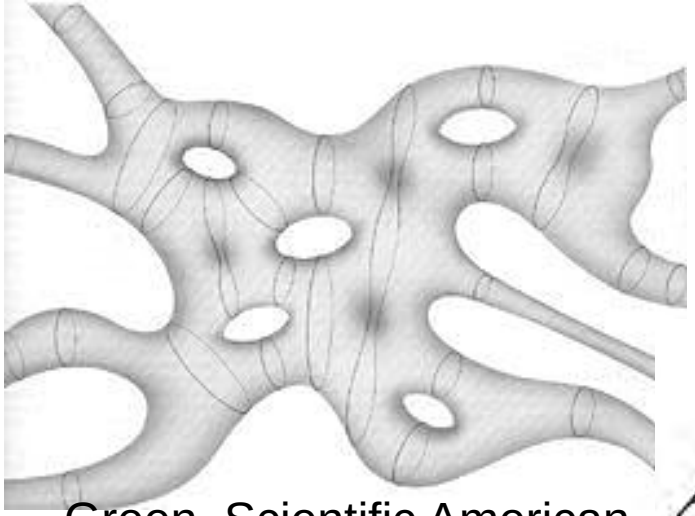
Thank you !!!

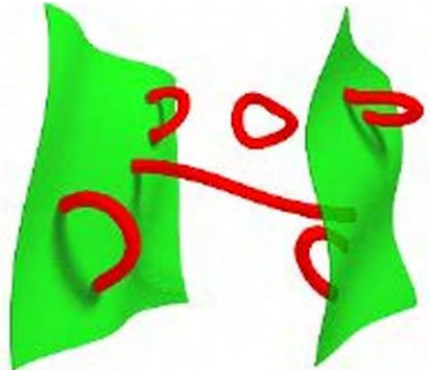


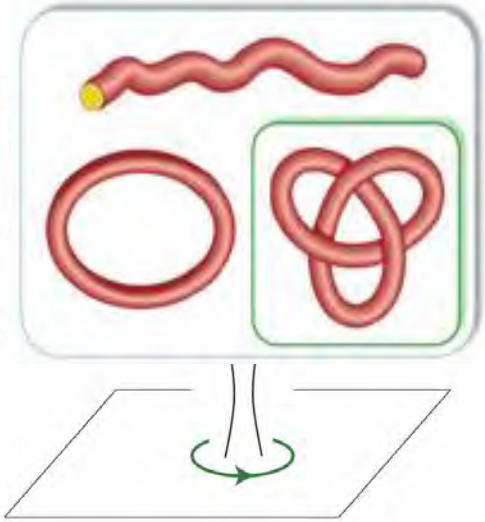
Topology in Different Fields of Science

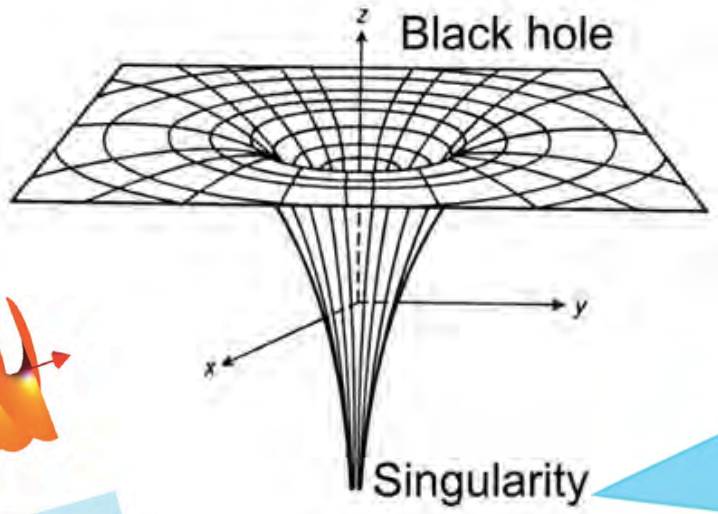
WORLD SHEET swept out by stringlike particles as they move in spacetime

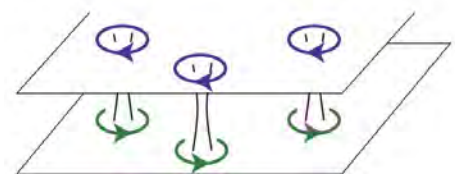


Green, Scientific American

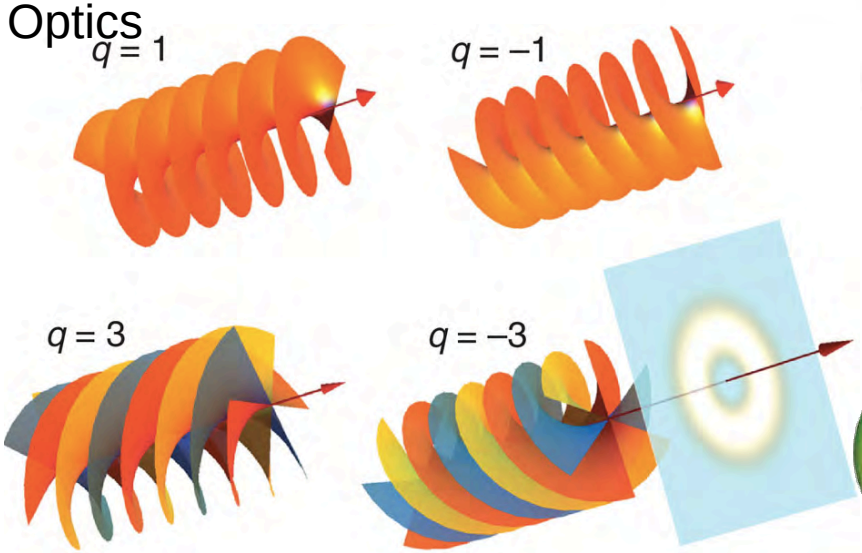


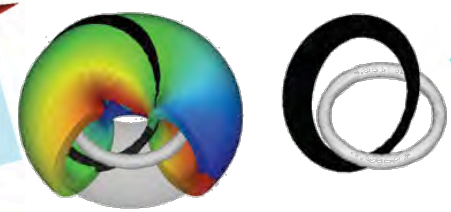


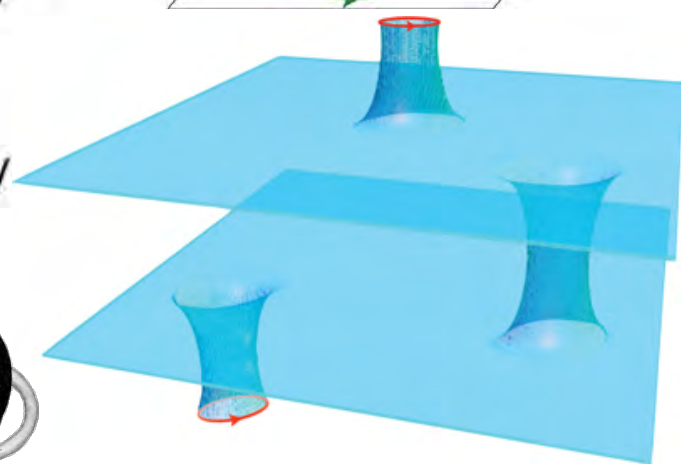




Optics







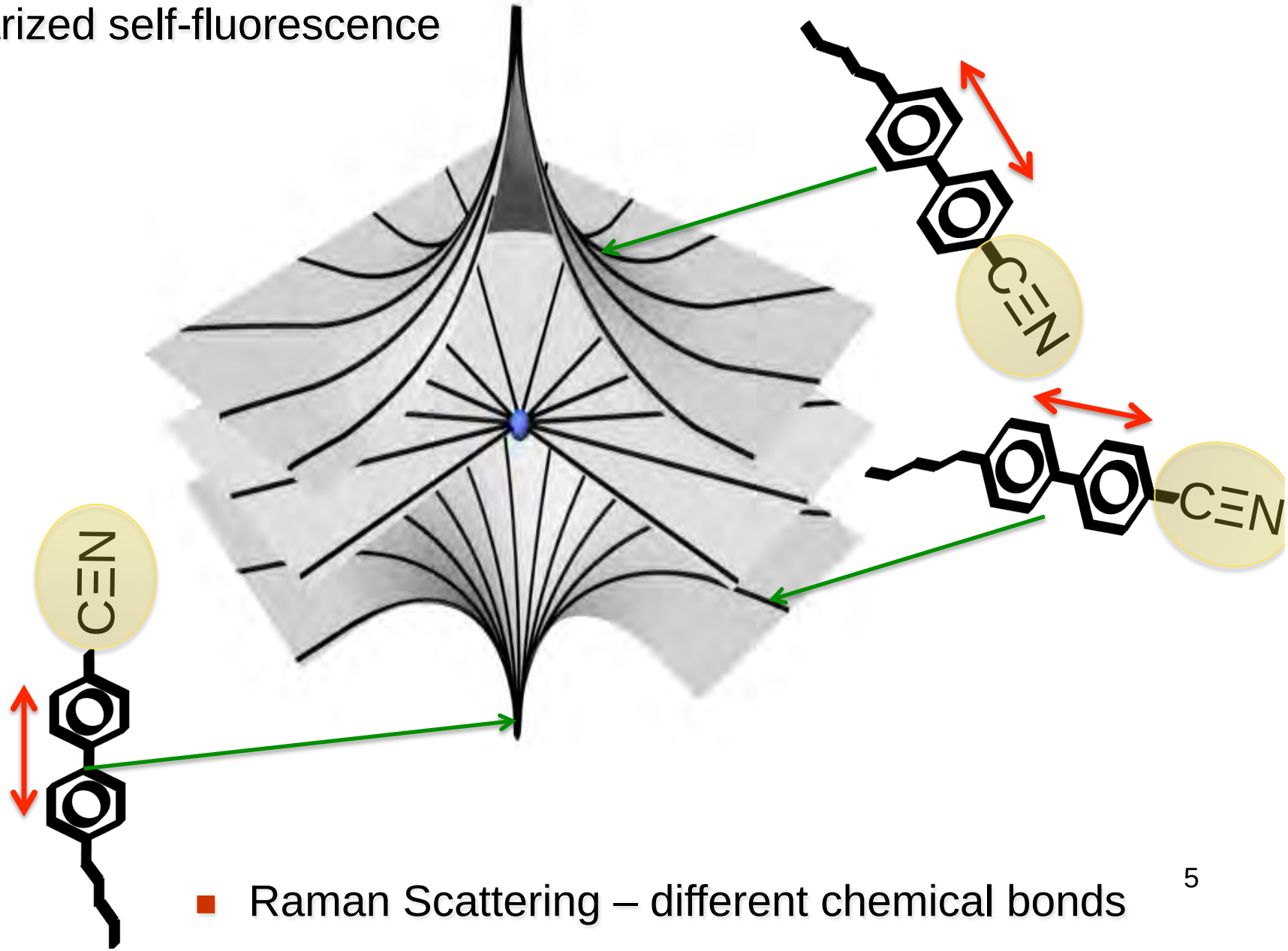
Topology and Soft Matter

Intro: Imaging, manipulation, fabrication, & aspects of topology

- Interplay of topologies of surfaces, fields & defects
- 2D “baby” skyrmions & 3D knotted solitons
- Soft matter defects & optical vortices
- Self-assembly & dynamics of knot solitons

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

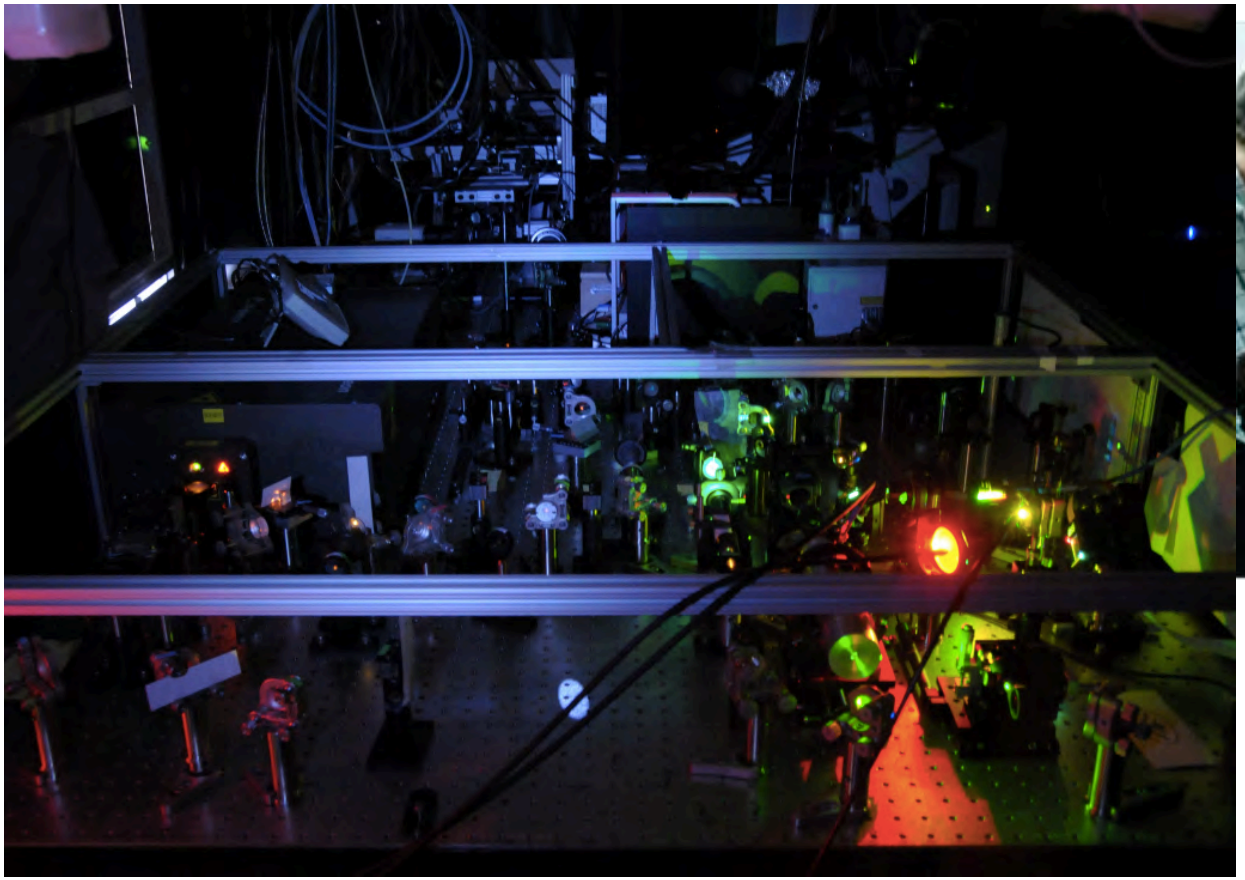
- Polarized self-fluorescence



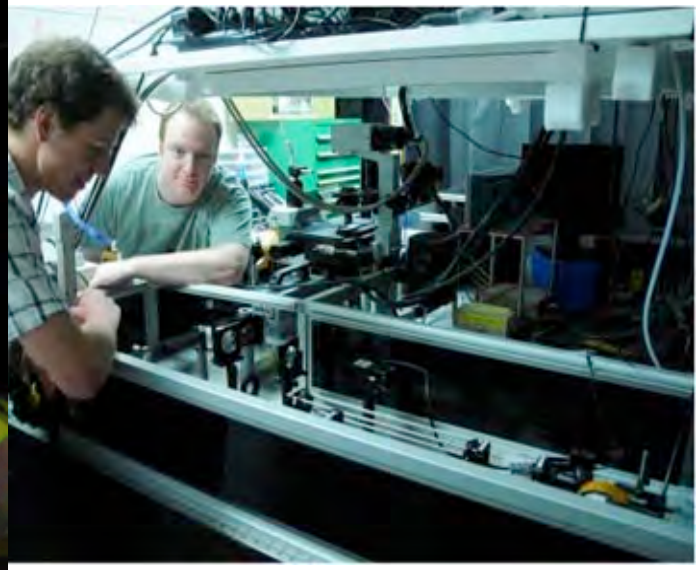
- Raman Scattering – different chemical bonds

Multimodal nonlinear optical imaging of LCs

→3D linear/nonlinear optical imaging



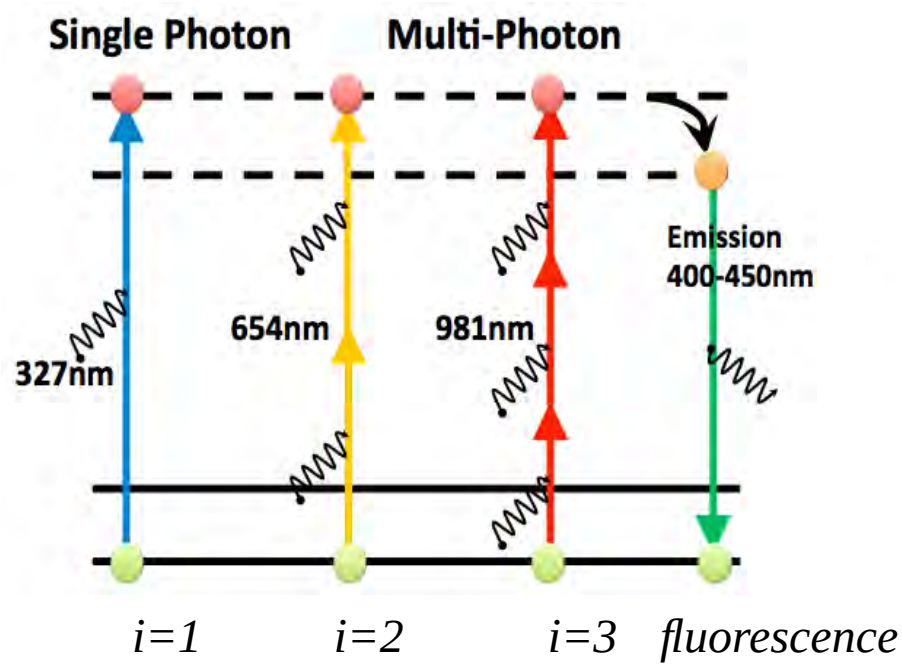
→3D optical manipulation

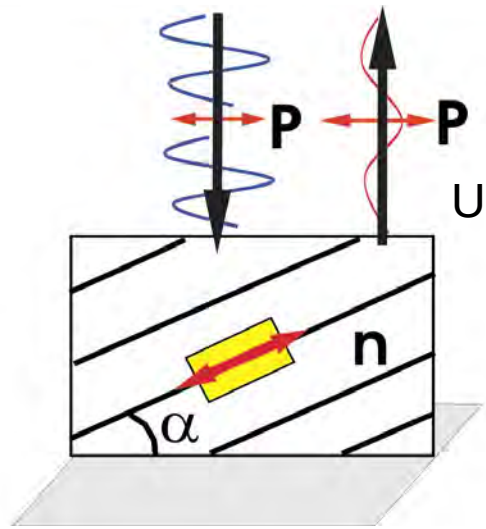


→Broadband CARS-PM
→Broadband SRS-PM
→Multiphoton fluorescence
→Multiple harmonic generation
→Holographic Optical Tweezers

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

Multiphoton self-fluorescence imaging: no dyes





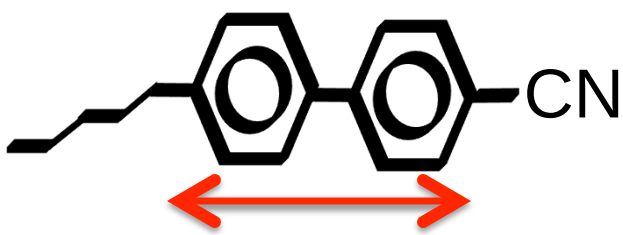
Unpolarized detection:

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

Polarized detection:

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

5CB

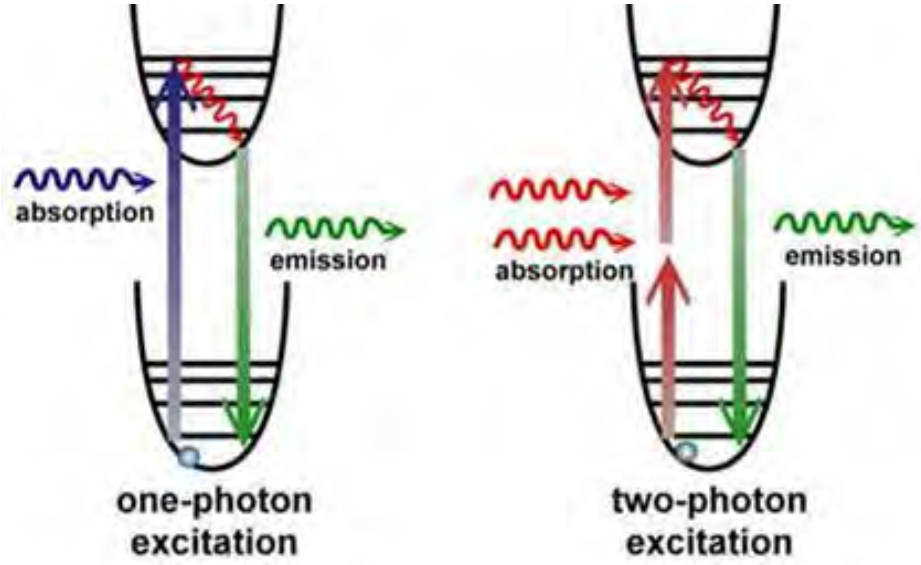


Absorption/emission transition dipole

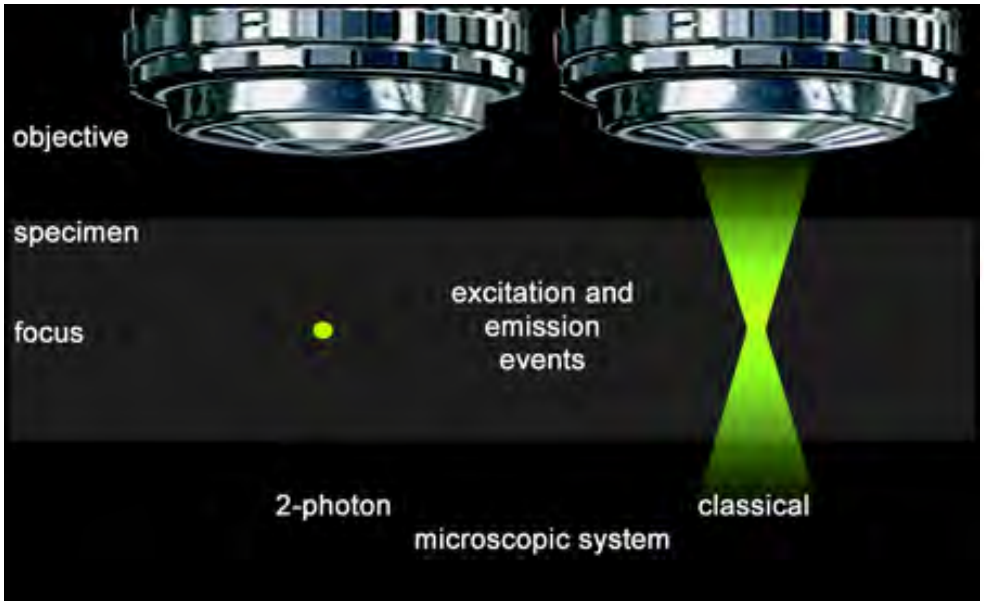
- **Strong orientational sensitivity!**
- **No dyes needed!!!**

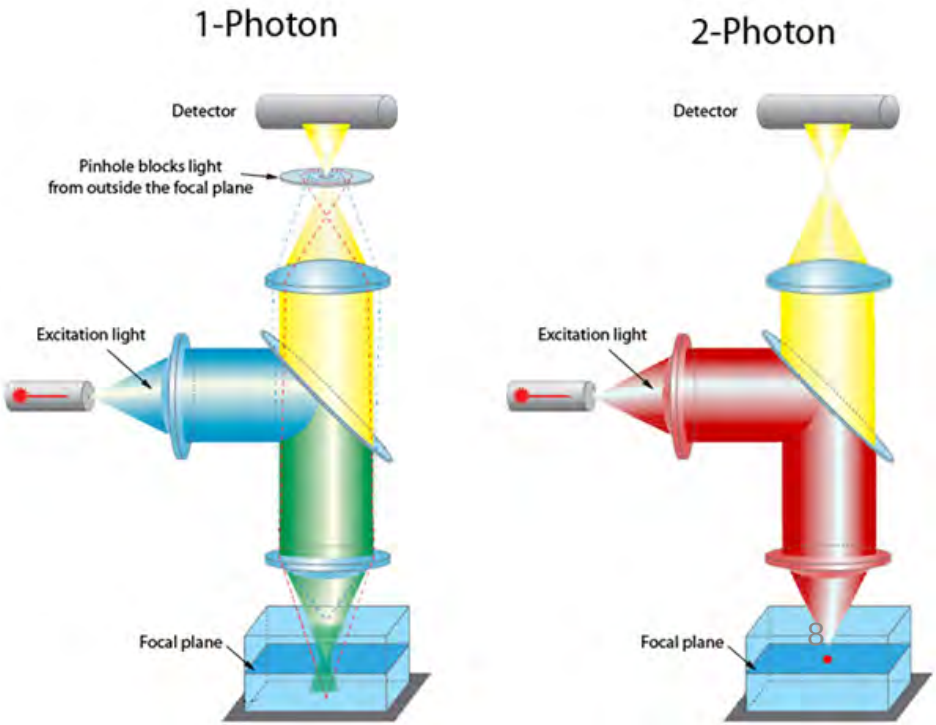
T. Lee, R.P. Trivedi, & I.I. Smalyukh, *Opt. Lett.* 35, 3447-3449 (2010).

Single- vs. Multi-photon Excitation



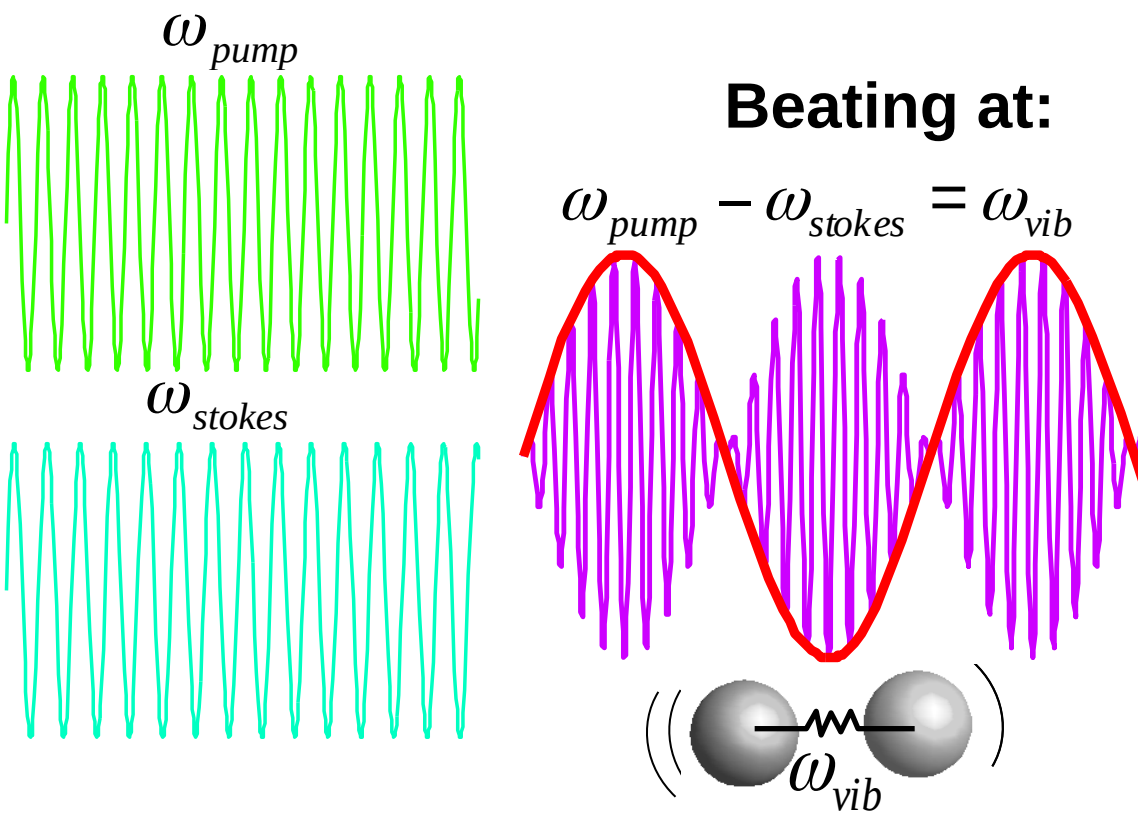
- **probability of simultaneous absorption of two or more photons is very low**
- **intensity of the fluorescence light falls off very fast above and below focus**
- **Intrinsic 3D resolution - no pinhole needed**

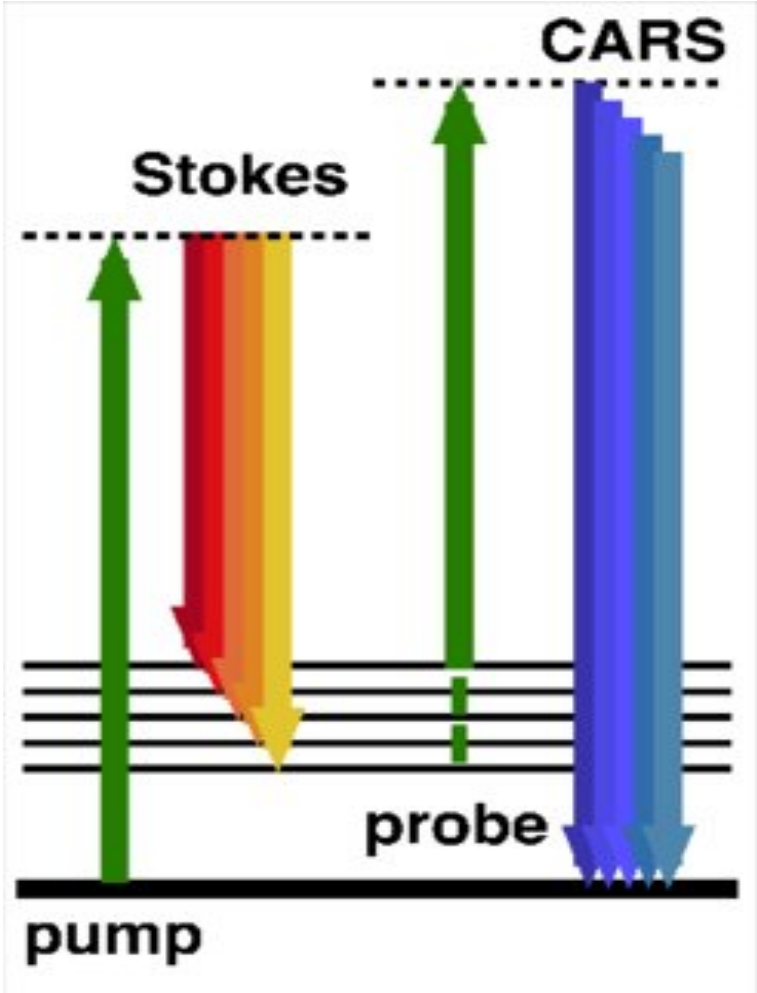




Coherent anti-Stokes Raman Scattering (CARS)

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



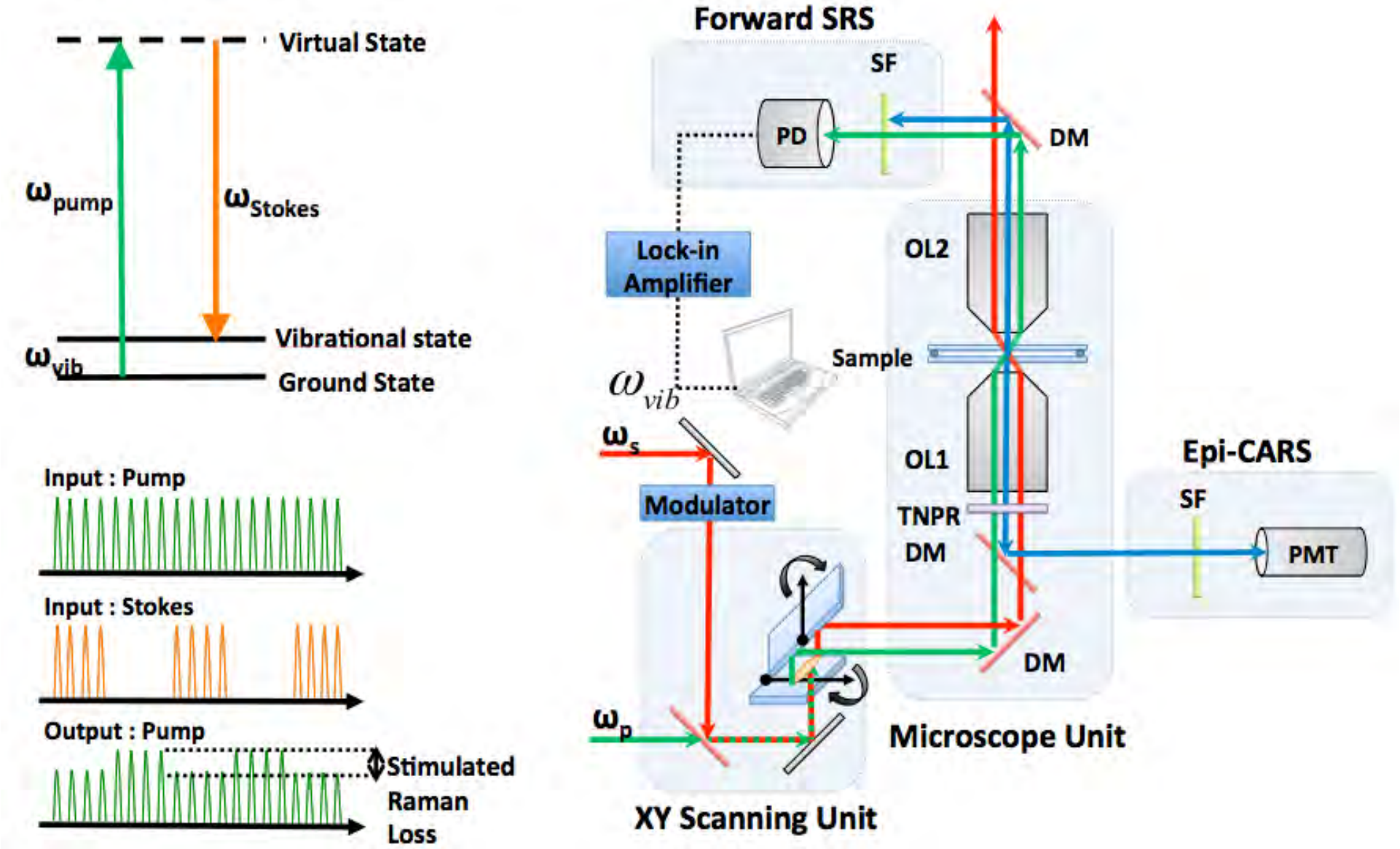


→ 3rd order nonlinear process

→ Beat frequency matches that of a Raman vibration -> signal at $\omega_{as}=2\,\omega_p - \omega_S$

→ Broadband Stokes excitation & CARS detection with Spectral shaping;

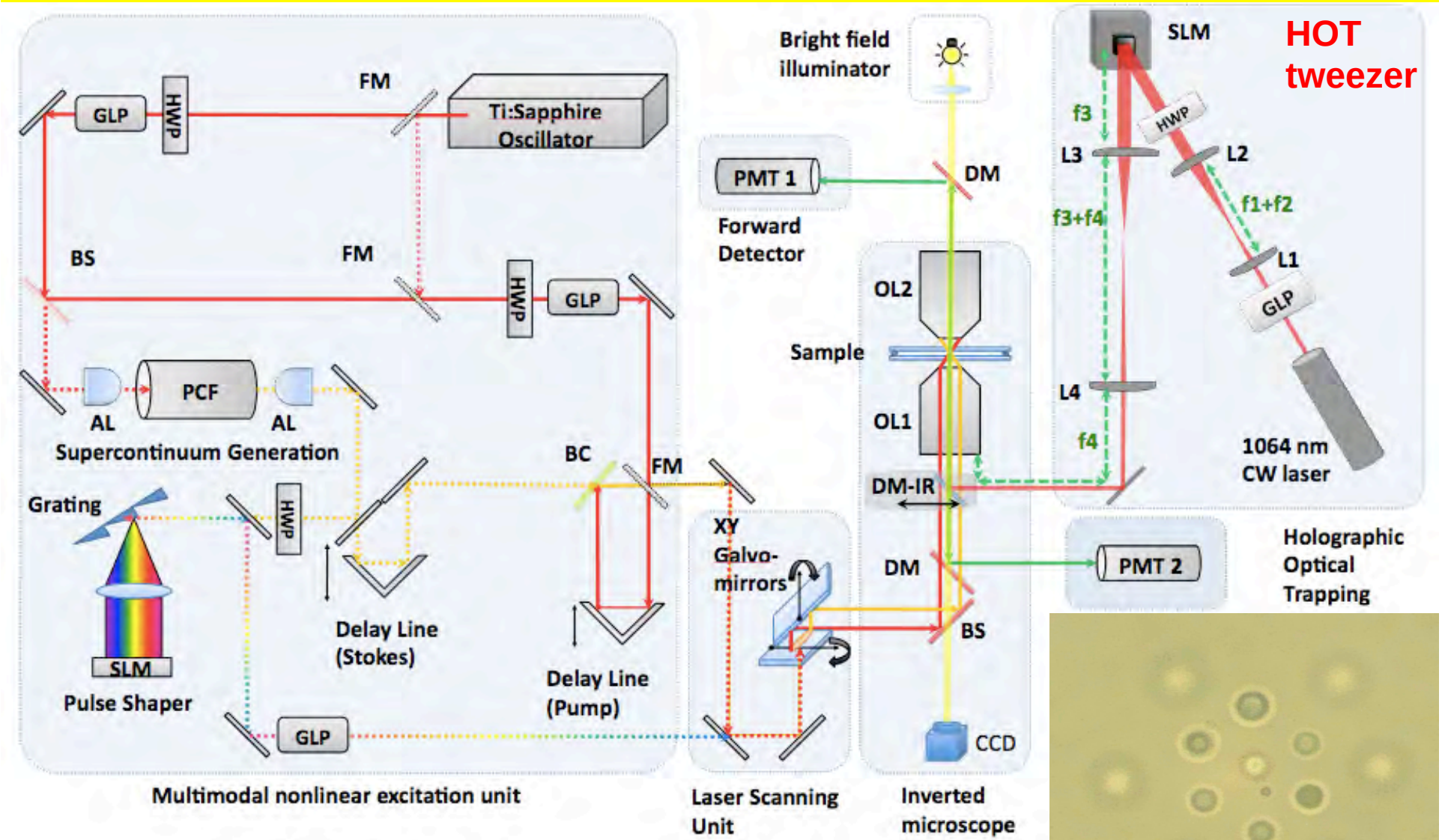
Stimulated Raman scattering polarizing m-py



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

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

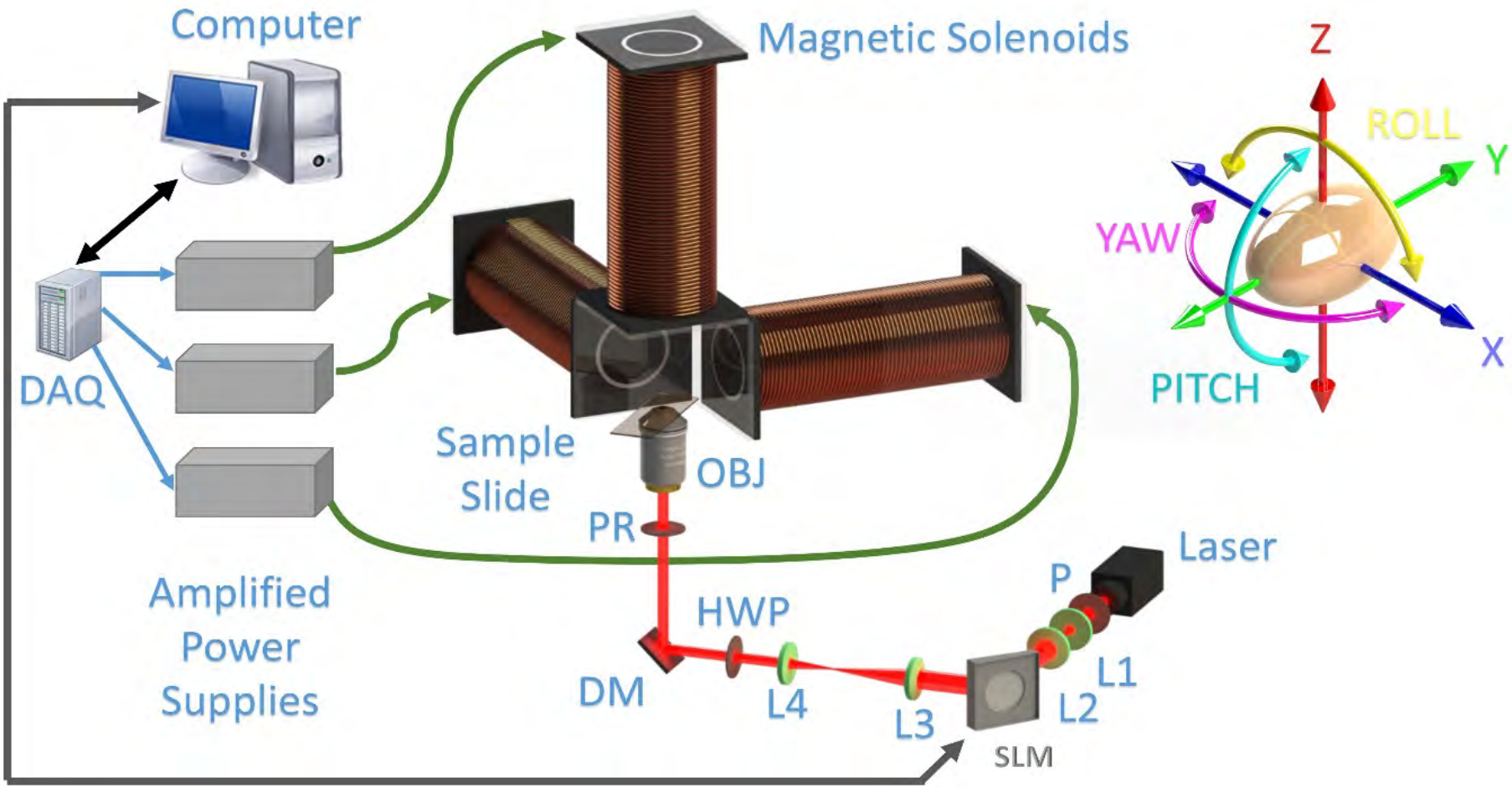
Integrated holographic optical tweezers & multimodal 3D imaging: setup schematic



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

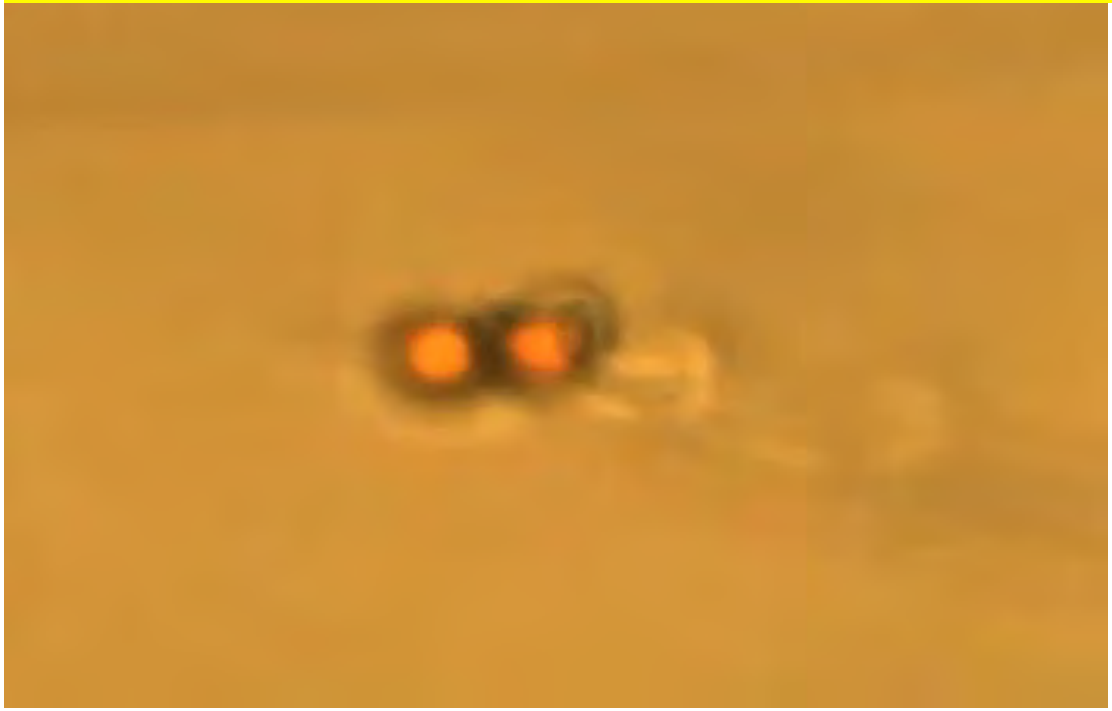
R.P. Trivedi, T. Lee, K. Bertness, & I.I. Smalyukh, *Opt. Express* 18, 27658-27669 (2010).

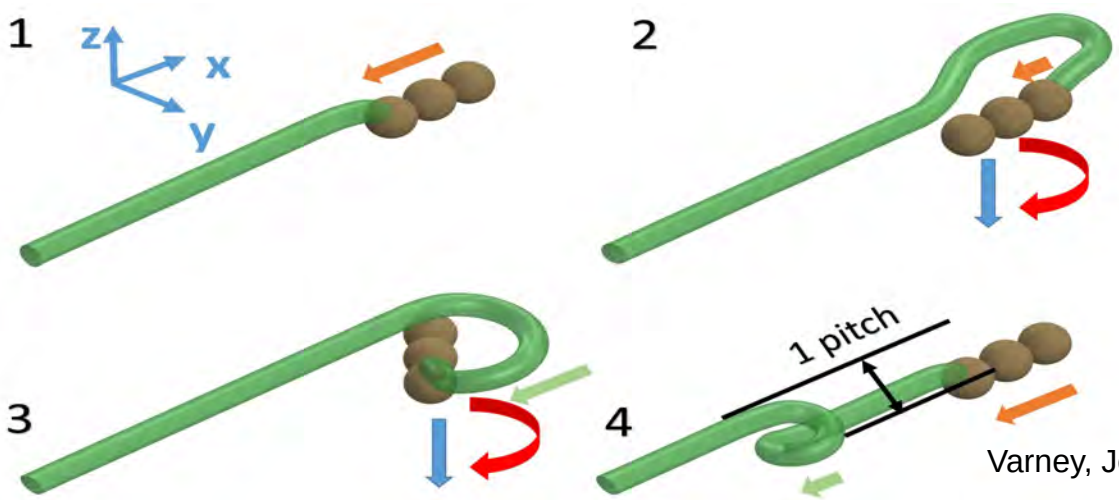
Magnetic & optical holonomic control



→Use colloidal objects with super-paramagnetic properties
→Control all degrees of freedom: rotational & translational

Controlling defects through colloidal handles





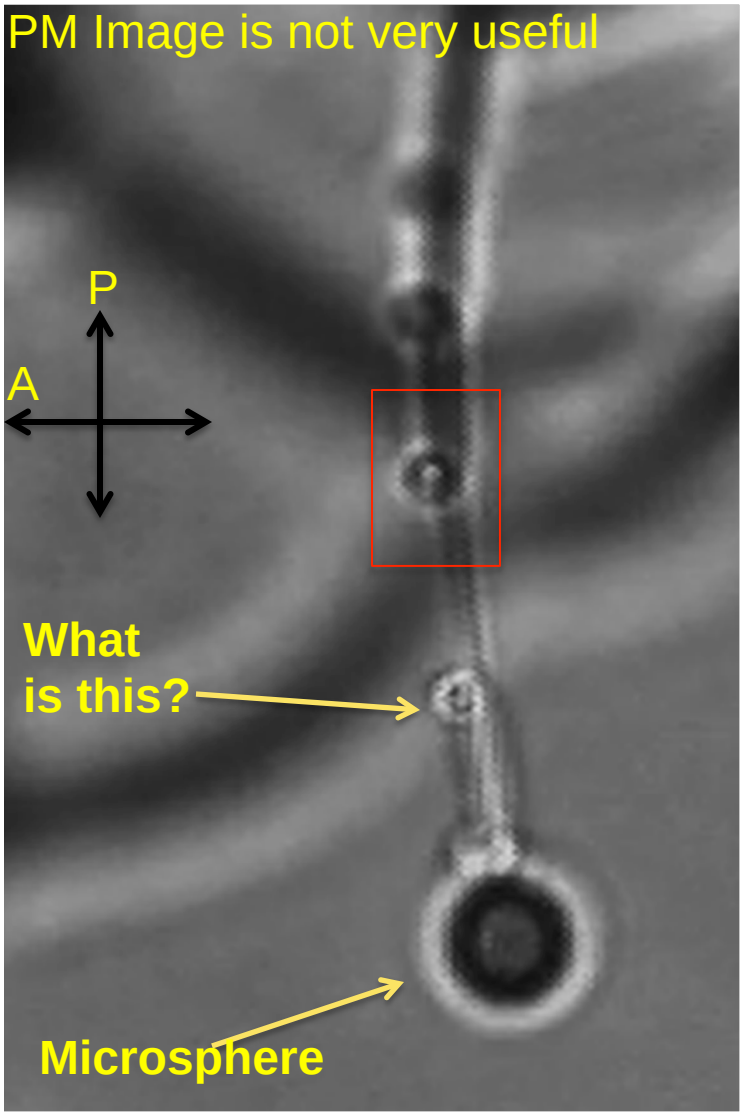
→Colloidal dimer of the super-paramagnetic beads

→What is the defect line?

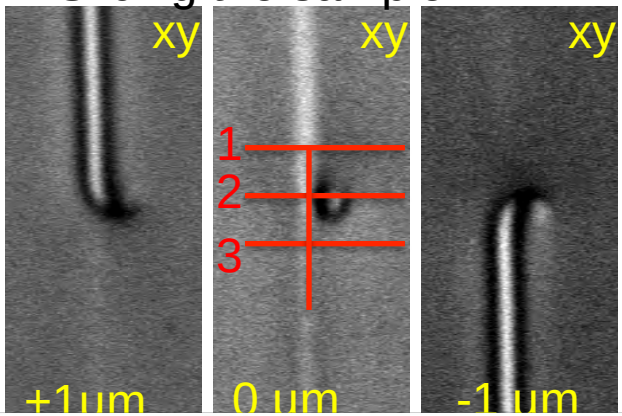
Varney, Jenness, Smalyukh. Phys Rev E **89**, 022505 (2014)

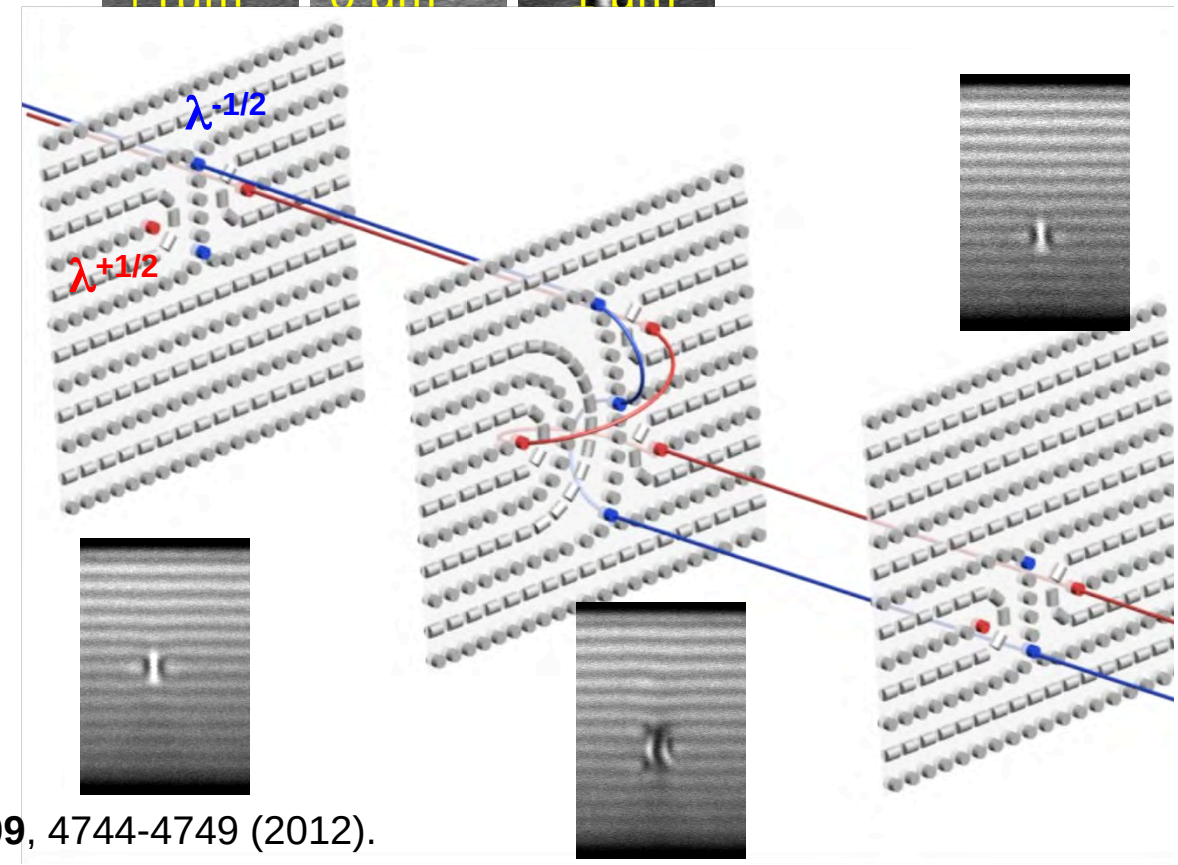
Simultaneous 3D "drawing" & imaging of defects

3D drawing of defects by a particle



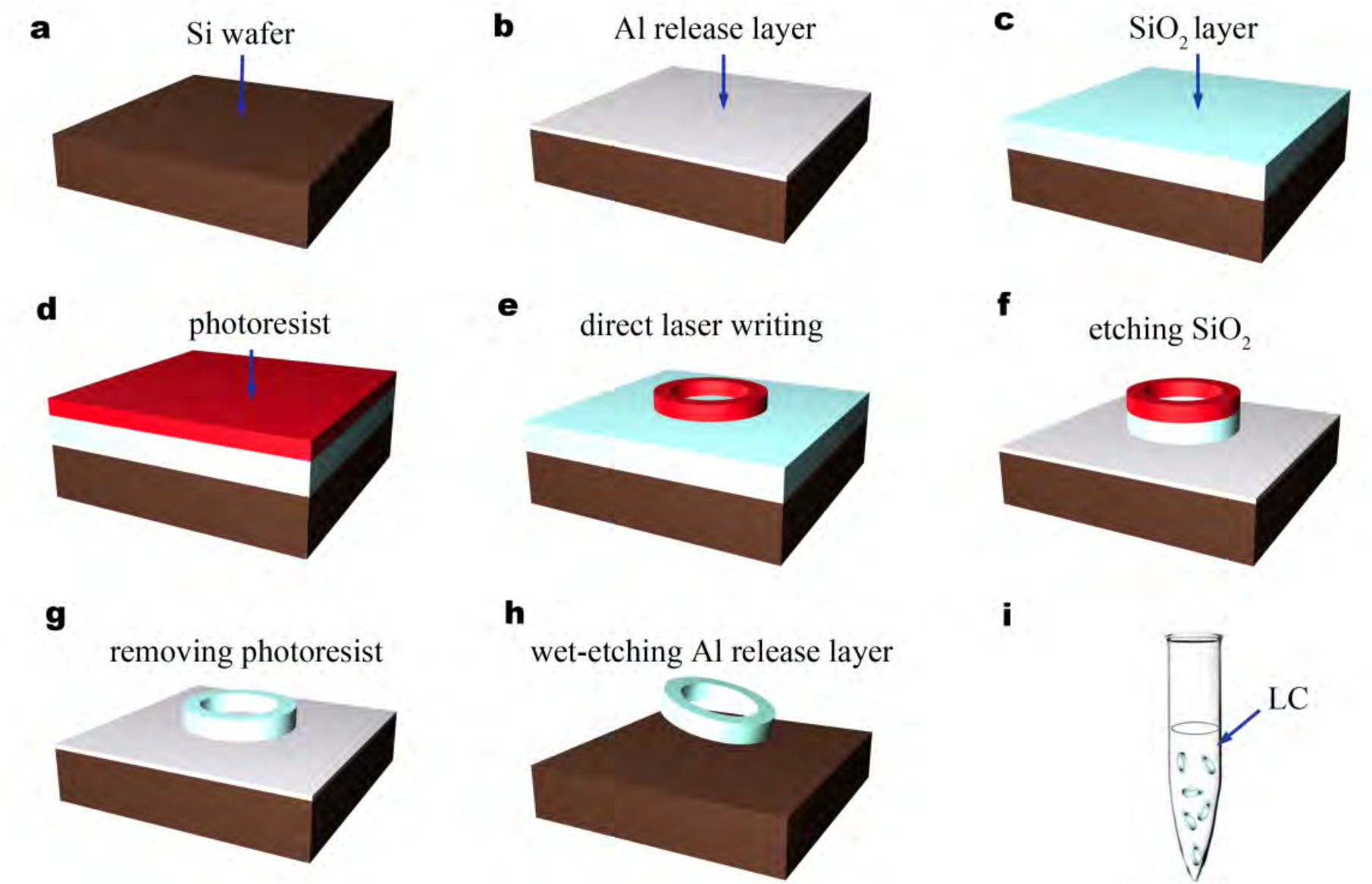
Slicing the sample





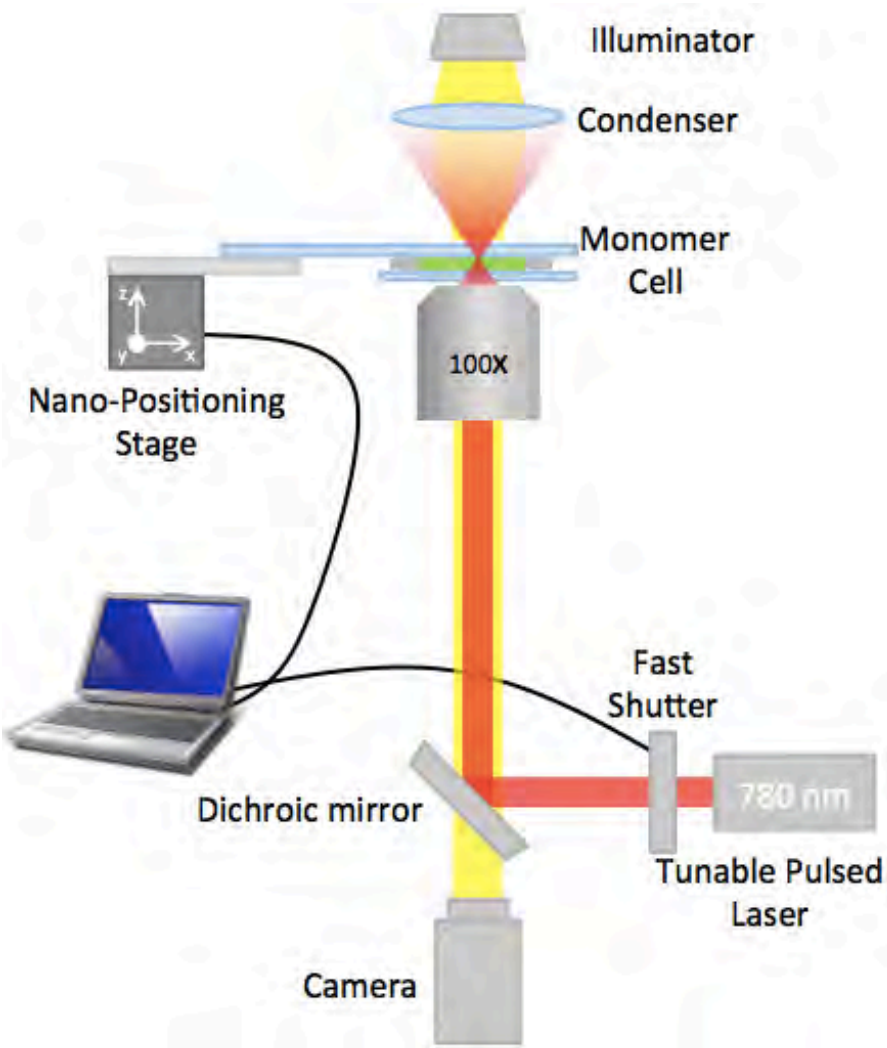
Trivedi, Senyuk, Lee, Smalyukh, PNAS **109**, 4744-4749 (2012).

Micro-fabrication of non-spherical colloids

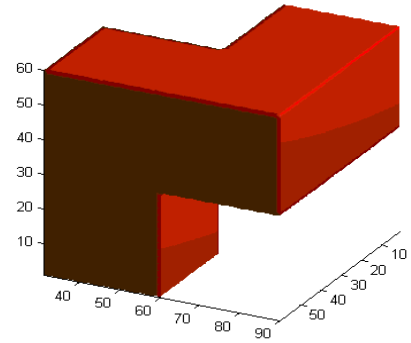


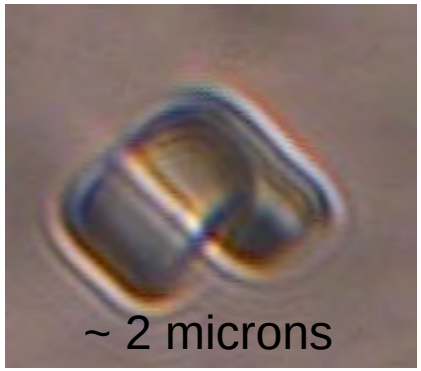
B. Senyuk, Q. Liu, S. He, R. Kamien, R. Kusner, T. Lubensky, & I.I. Smalyukh, Nature **493**, 200-205 (2013)

3D complex-shaped colloidal particles



- two-photon photopolymerization

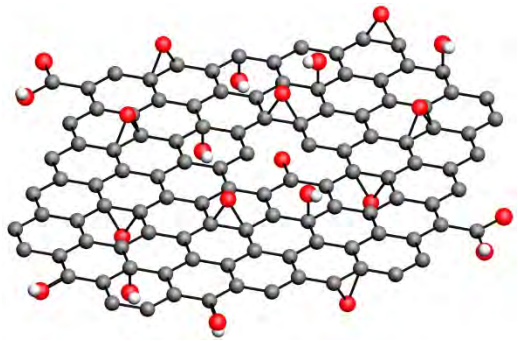




A. Martinez, T. Lee, T. Asavei, H. Rubinsztein-Dunlop, & I.I. Smalyukh, *Soft Matter* ***7****, 11154-11159 (2012).*

Aqueous Graphene Oxide Colloidal Liquid Crystals

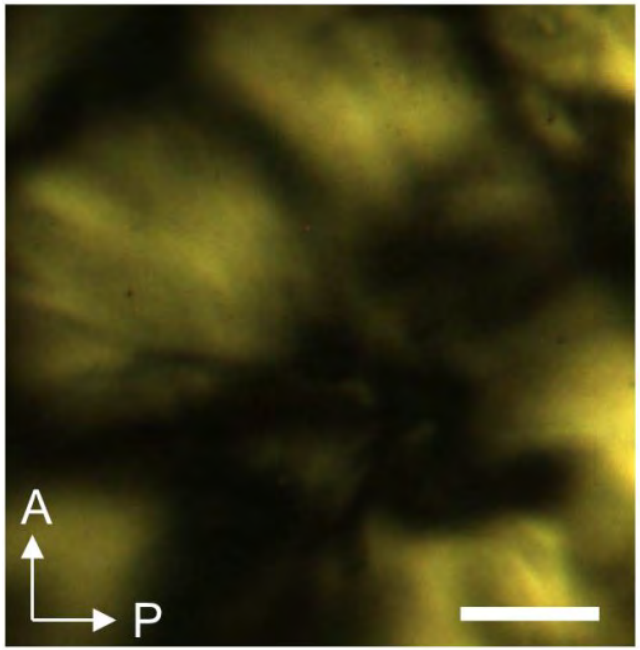
Aqueous GO flakes – 0.3-0.5 vol%, lateral size ~ 0.4 µm on average

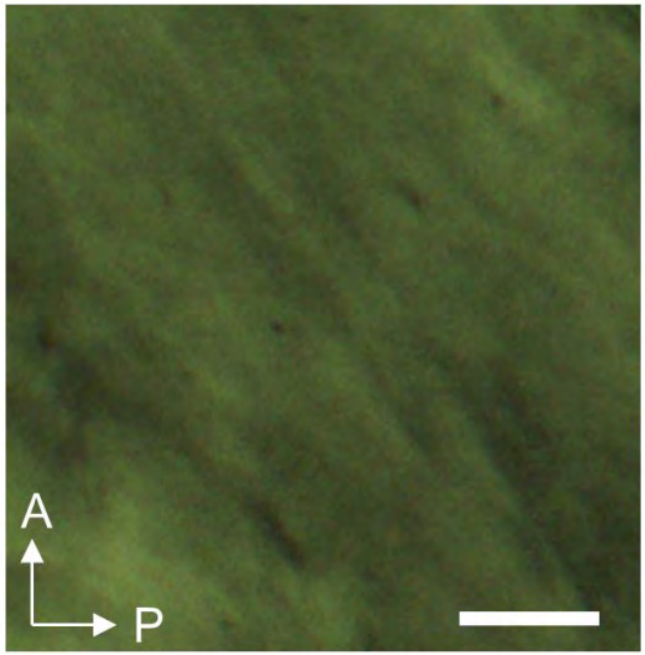


- Carbon
- Oxygen
- Hydrogen

→Electrostatic stabilization due to the charged groups!

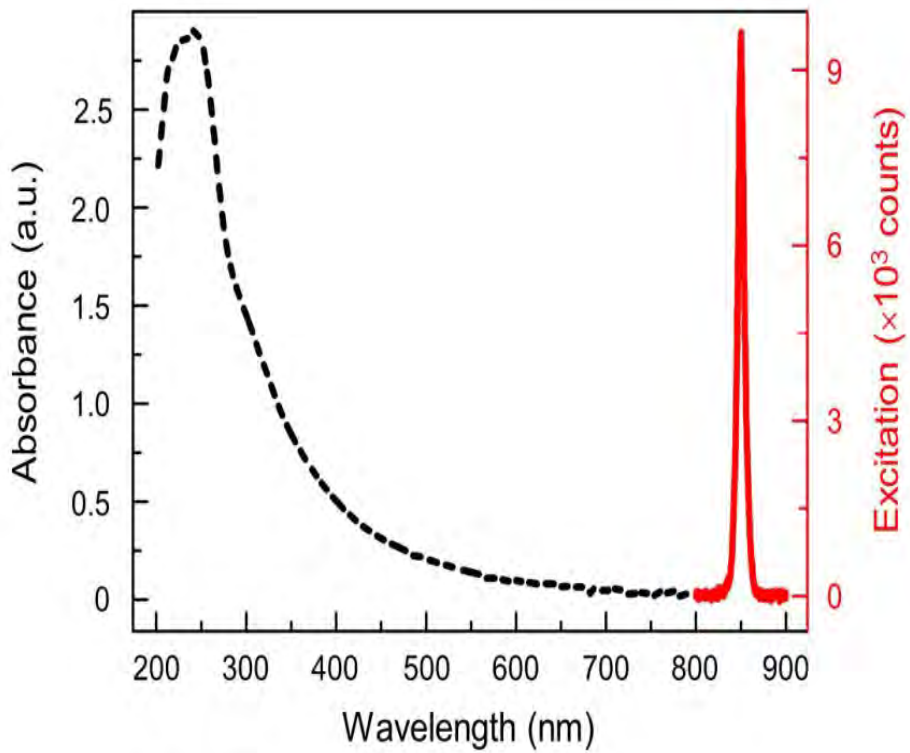
Textures of a GO nematic liquid crystal at concentration 0.7 wt. %

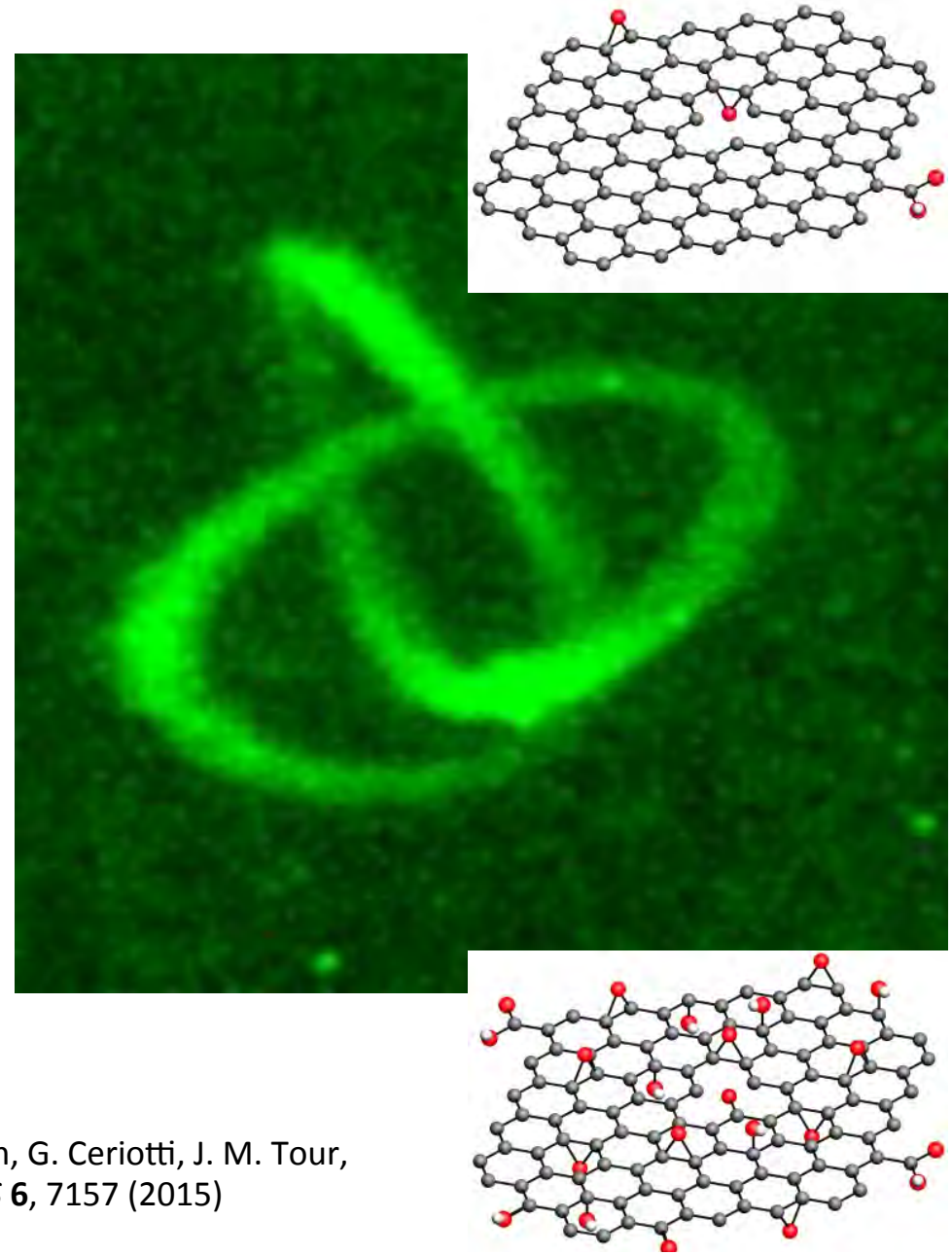




Defining complex-shaped colloidal microparticles

Optical properties of a GO nematic liquid crystal at concentration 0.7 wt. %

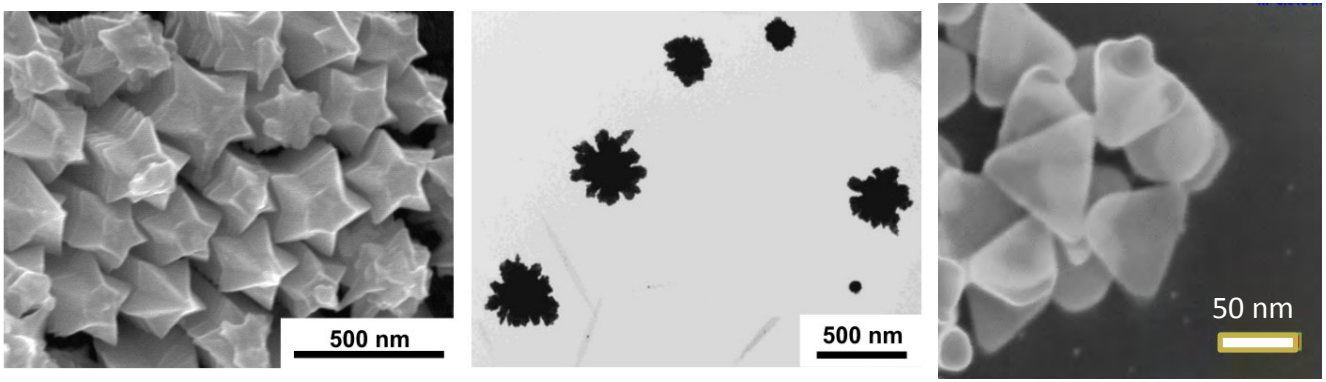




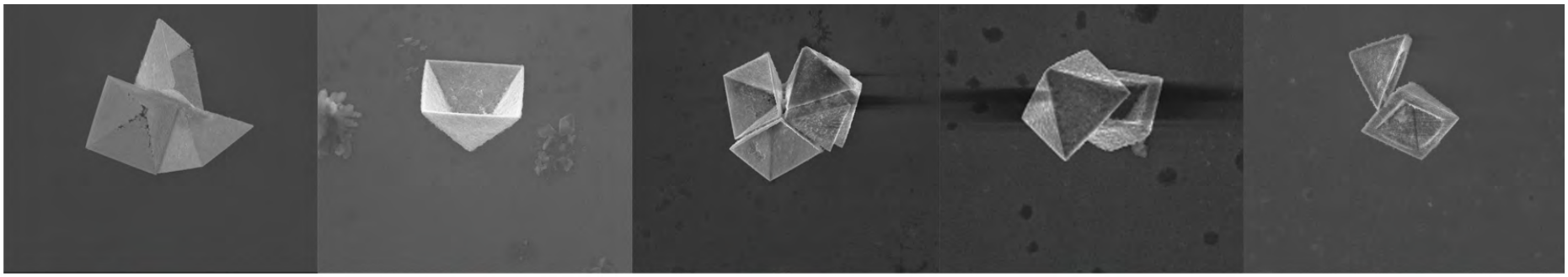
B. Senyuk, N. Behabtu, A. Martinez, T. Lee, D. E. Tsentalovich, G. Ceriotti, J. M. Tour, M. Pasquali, and I. I. Smalyukh. *NATURE COMMUNICATIONS* **6**, 7157 (2015)

Discussion/HW Assignment:

how would you fabricate such particles?



Colloidal particles shaped as hollow pyramids



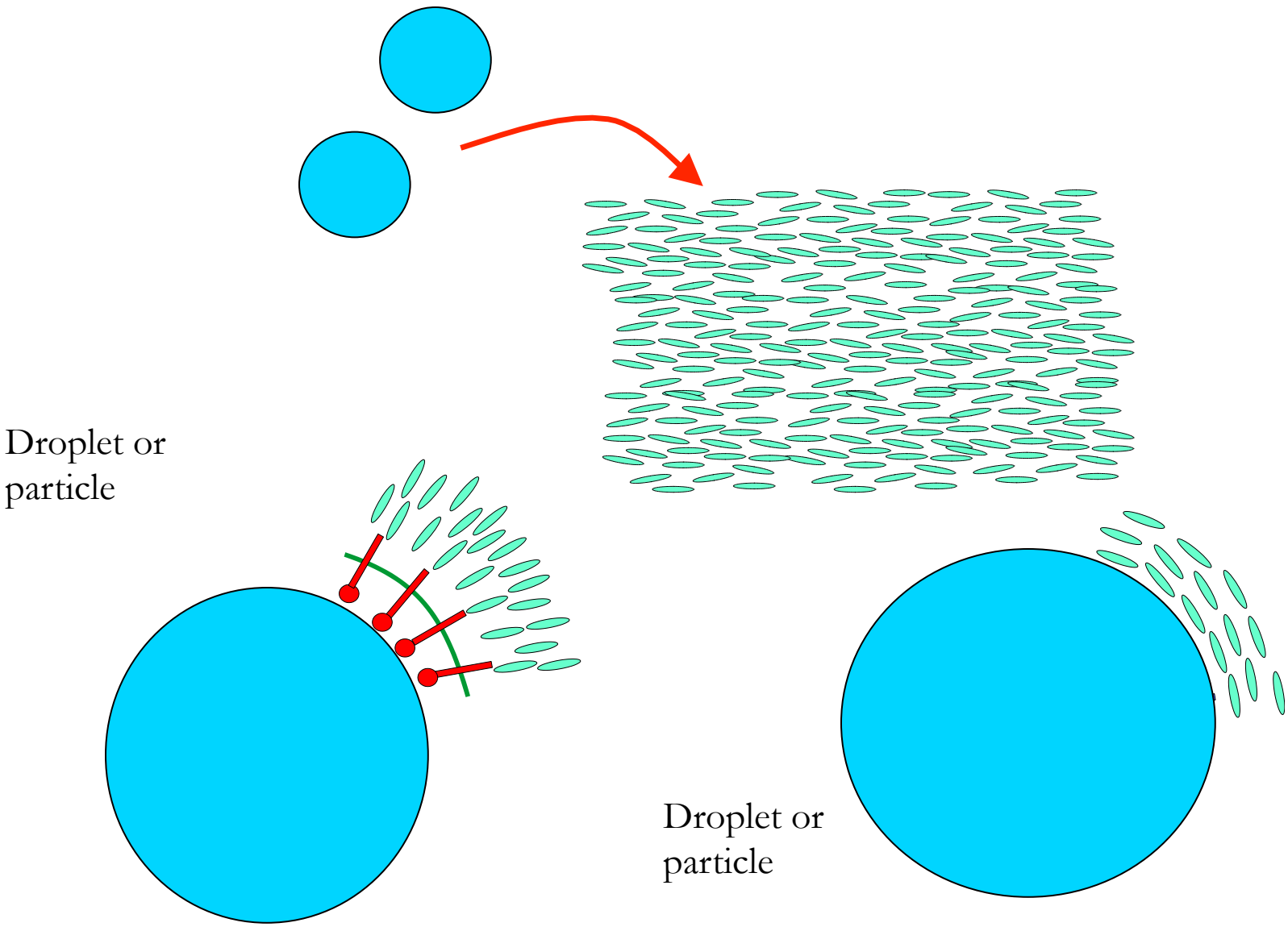
170nm 180nm 270nm 1000nm 1100nm

Tips:

- Note the overall size range provided in nanometers;
- Consider limitations of different techniques;
- Consider approaches not introduced here.

Interplay of topologies of fields, surfaces & defects probed using LC Colloids & Drops

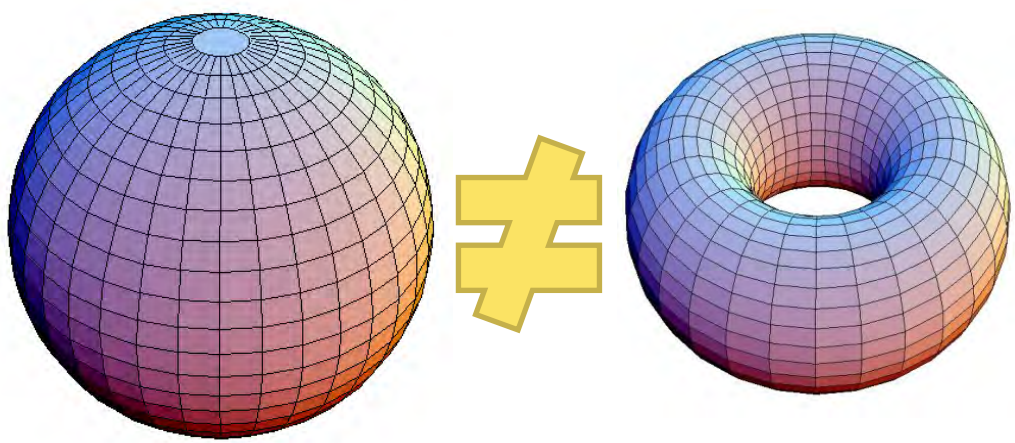
Inclusions/colloids in liquid crystals



Boundary conditions which drive the distortions

Topologically distinct colloidal surfaces?

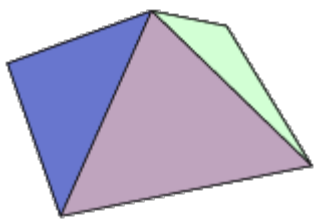
- Homeomorphic



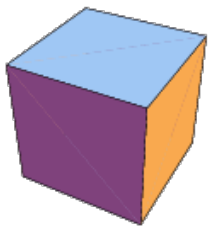
- Topologically distinct!

Topology of Surfaces: beyond spheres

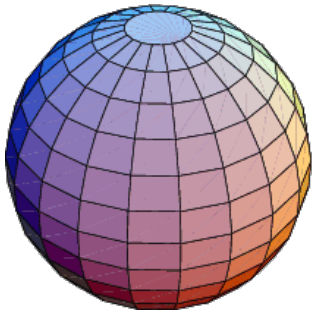
- Every surface can be assigned an Euler characteristic.
- For a geometry with F faces, V vertices, and E edges, the Euler characteristic is given by $\chi = V - E + F$.
- For an n-torus, the Euler characteristic is given by $\chi = 2 - 2n$.



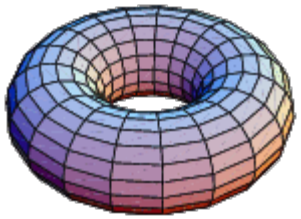
$\chi=2$



$\chi=2$



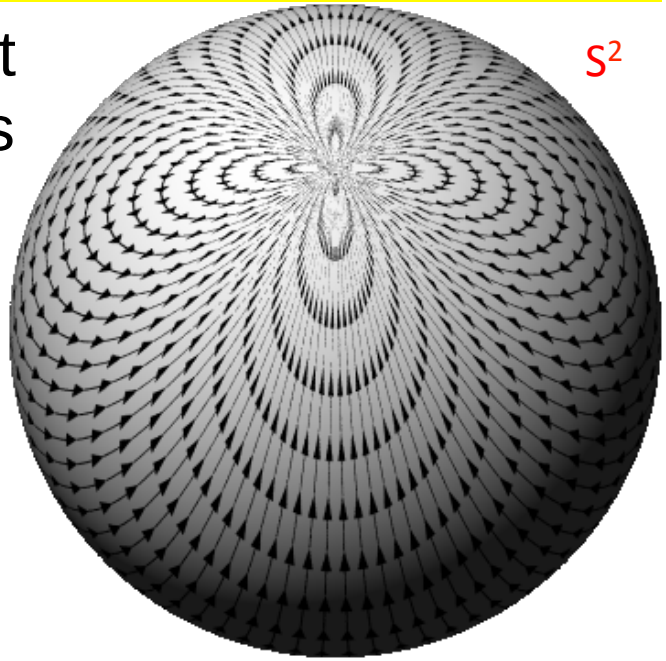
$\chi=2$



$\chi=0$

Hairy Ball & Poincare-Hopf theorems

Hairy Ball Theorem: no continuous tangent vector field on even-dimensional S^n-spheres (e.g. S^2)



– discussed previously in relation to spherical droplets & particles

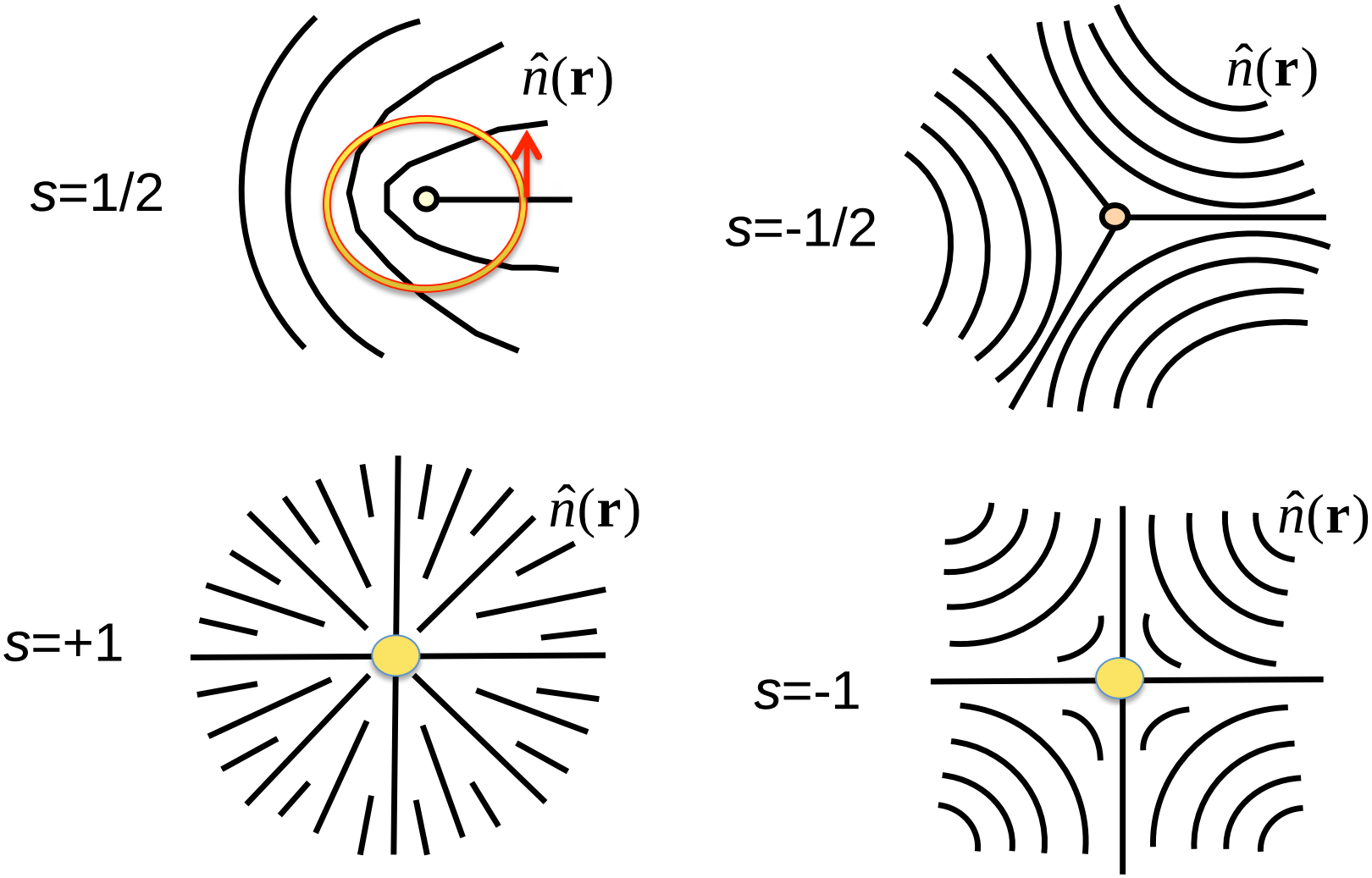
Poincare-Hopf theorem: winding numbers of defects in the vector field add to the Euler characteristic: $\chi = 2 - 2g$

$g = 0$ $\quad$ $g = 1$ $\quad$ $g = 2$ $\quad$ $g = 3$

$\chi = 2$ $\quad$ $\chi = 0$ $\quad$ $\chi = -2$ $\quad$ $\chi = -4$

. . .

Defects in 2D: Winding Number



- Director rotates by $2\pi s$ on going around it (s is the winding number)
- Recall this is different in 3D (integer unstable...)

Topology-required surface defects - boojums

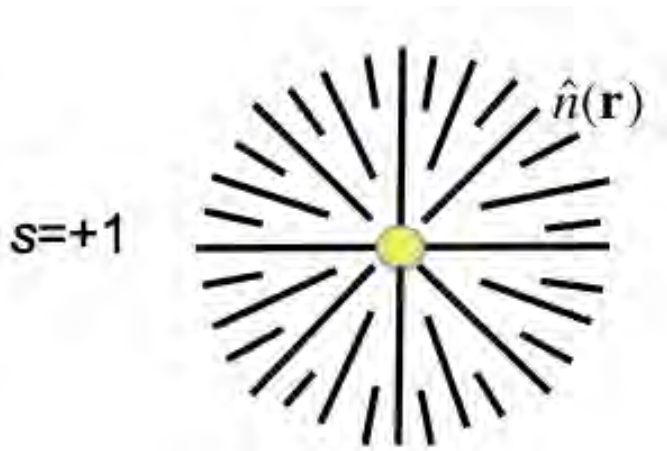
Spherical particle with tangential surface BCs

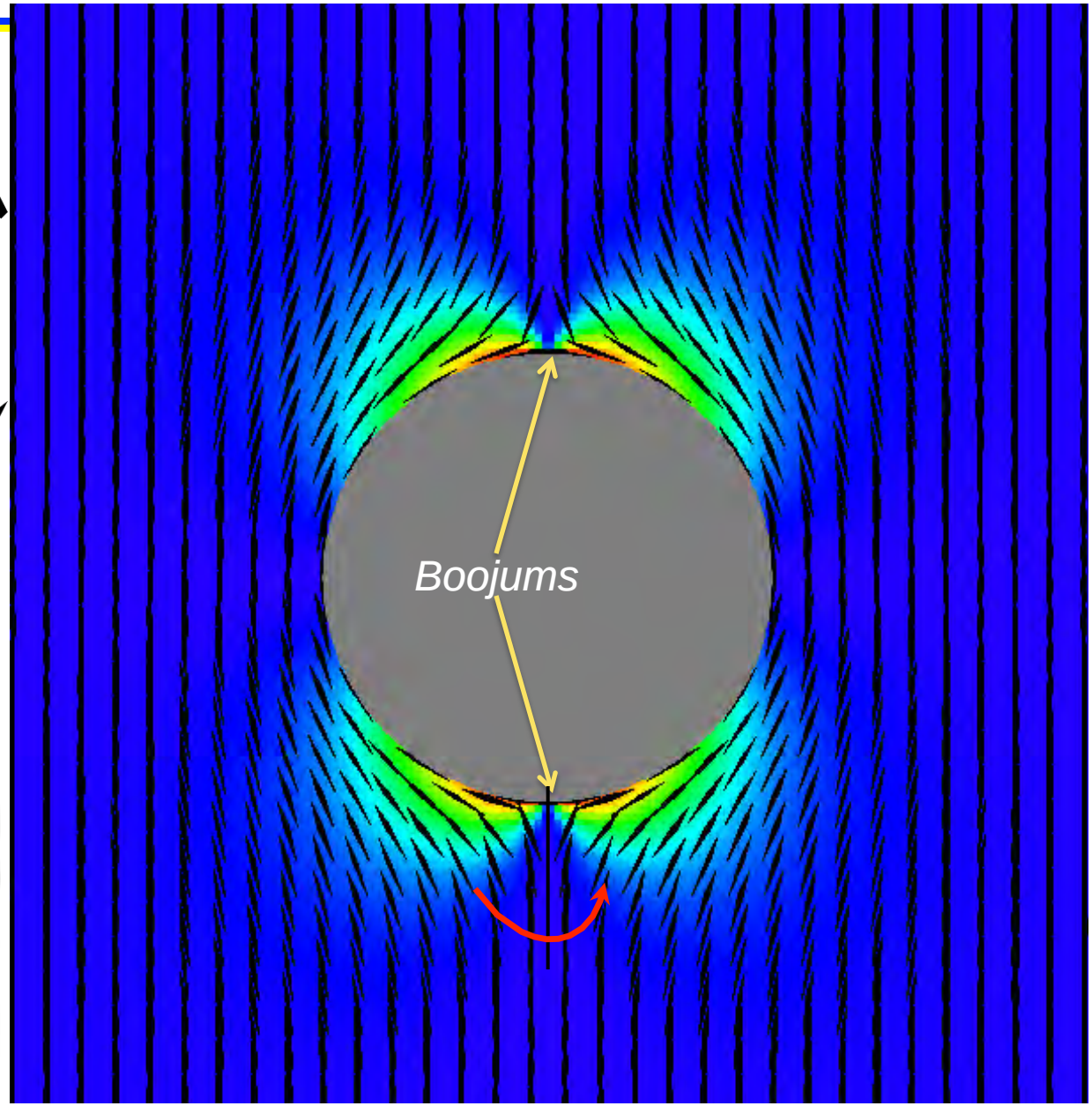
Two boojums at poles

$\hat{n}_0$

Winding number of defects in the 2D field at the LC particle interface is

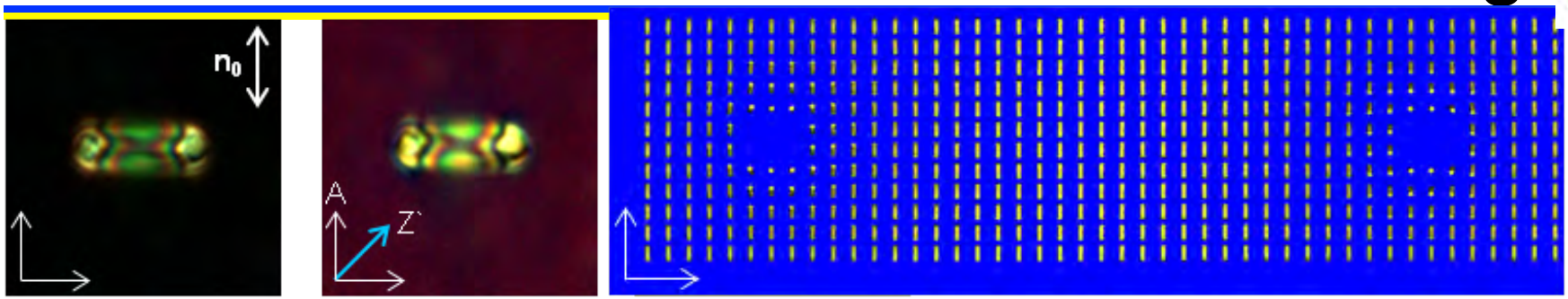
$$s = \chi = 2$$

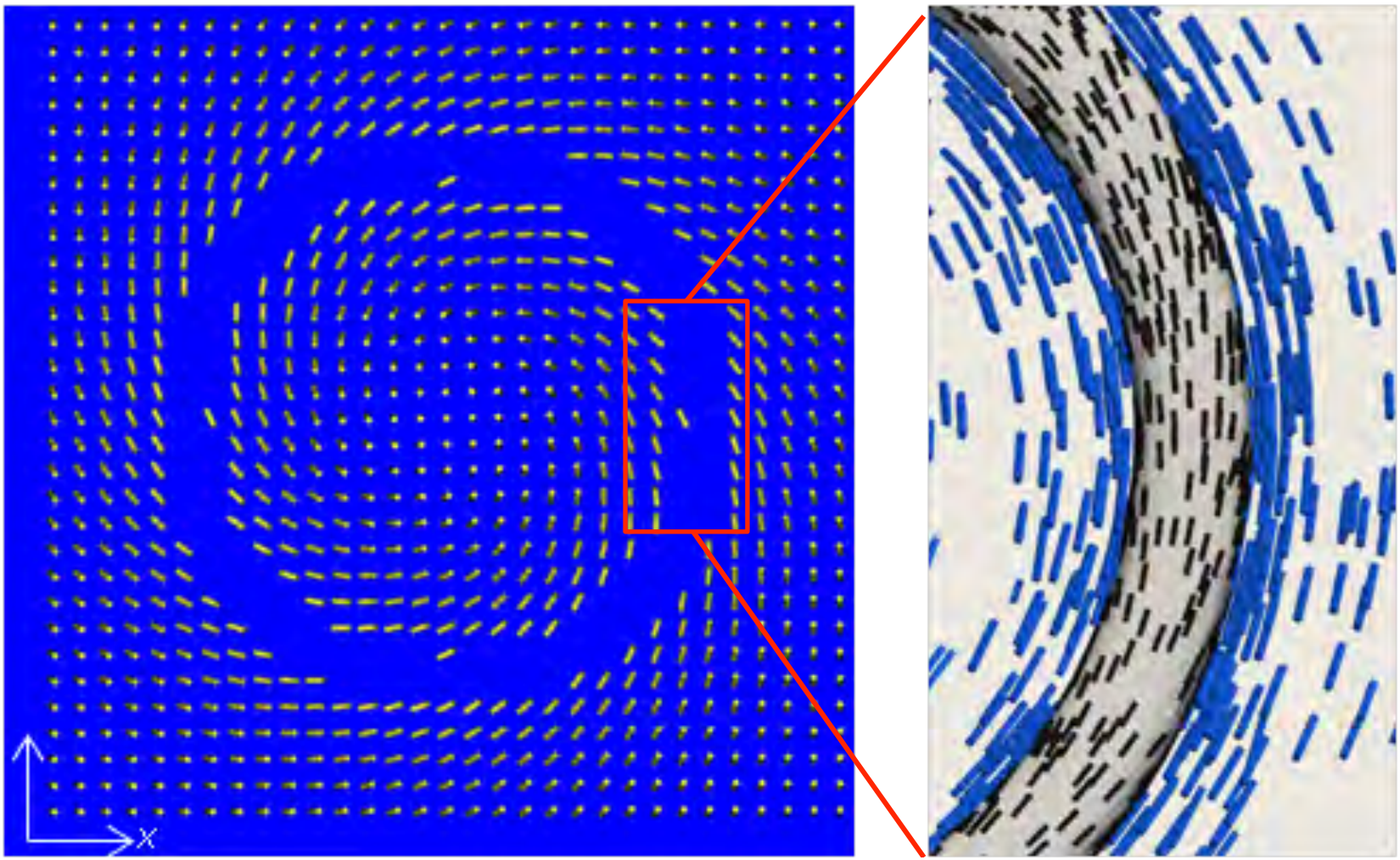




Proc. Natl. Acad. Sci. U.S.A. **103**, 18048 (2006)

Defect-free structures around a ring

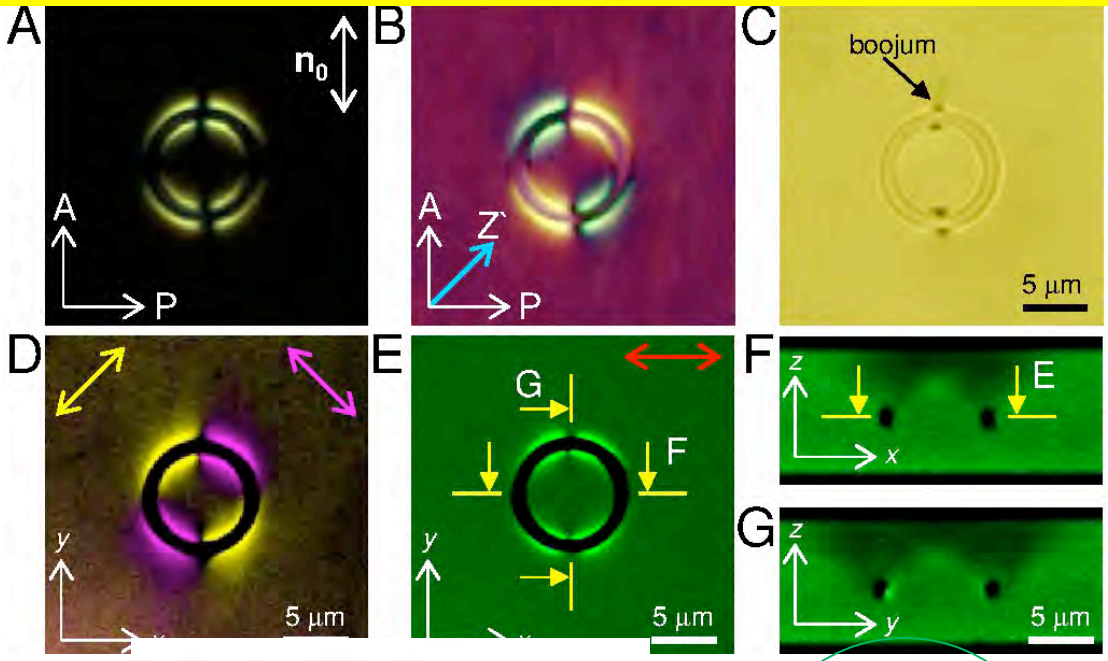


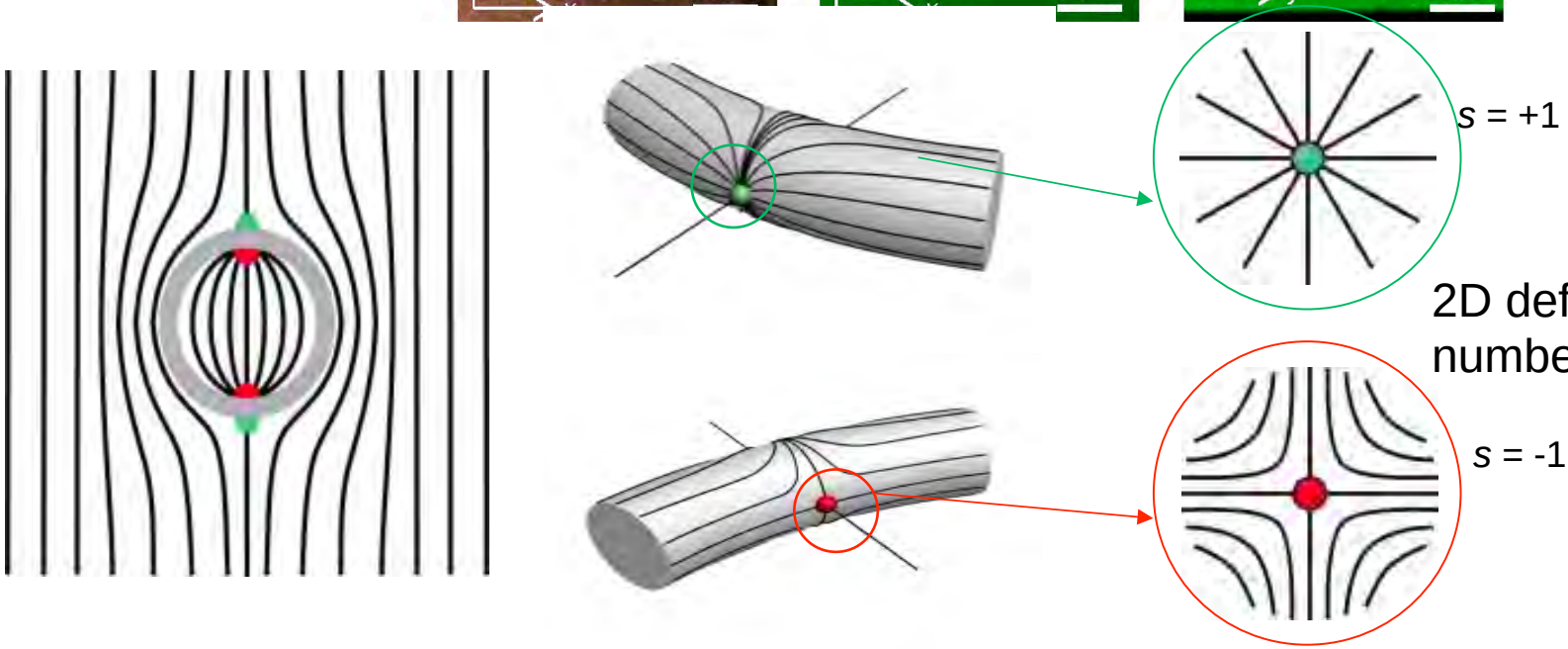


Boojums on colloidal rings

Polarizing optical microscopy

3D imaging





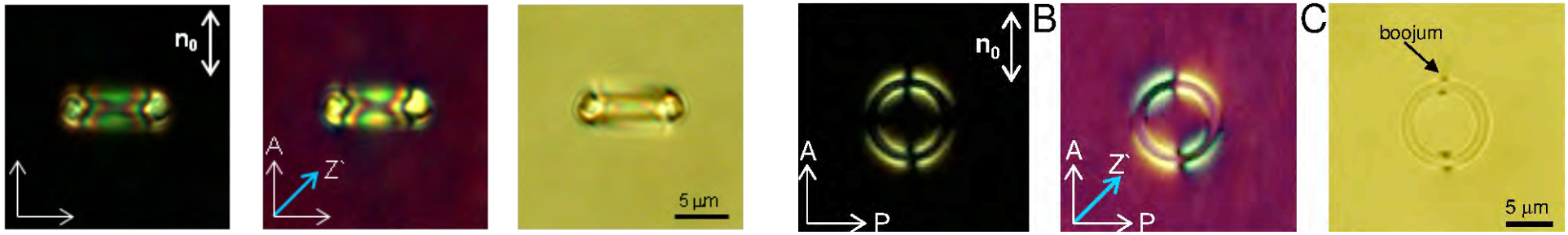
2D defect winding numbers

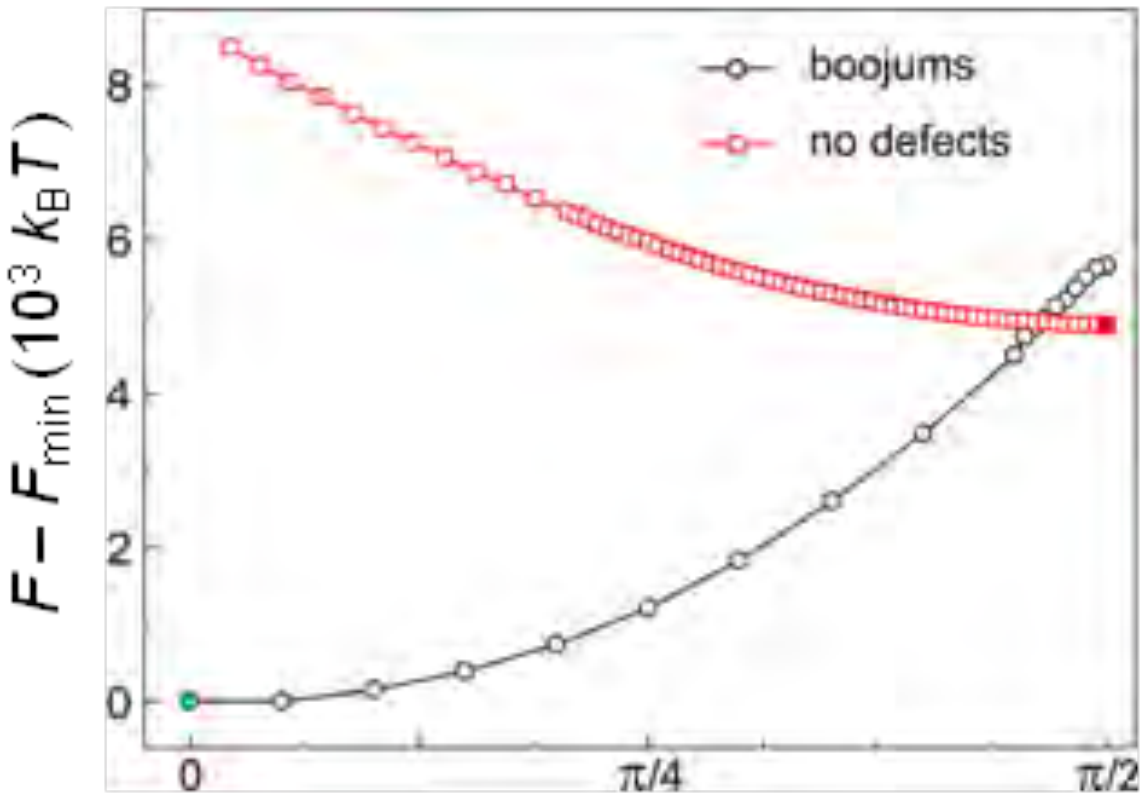
Liu, Senyuk, Tasinkevych, Smalyukh. *Proc. Natl. Acad. Sci. U.S.A.* **110**, 9231 (2013)

Structural stability

No defects

With boojums

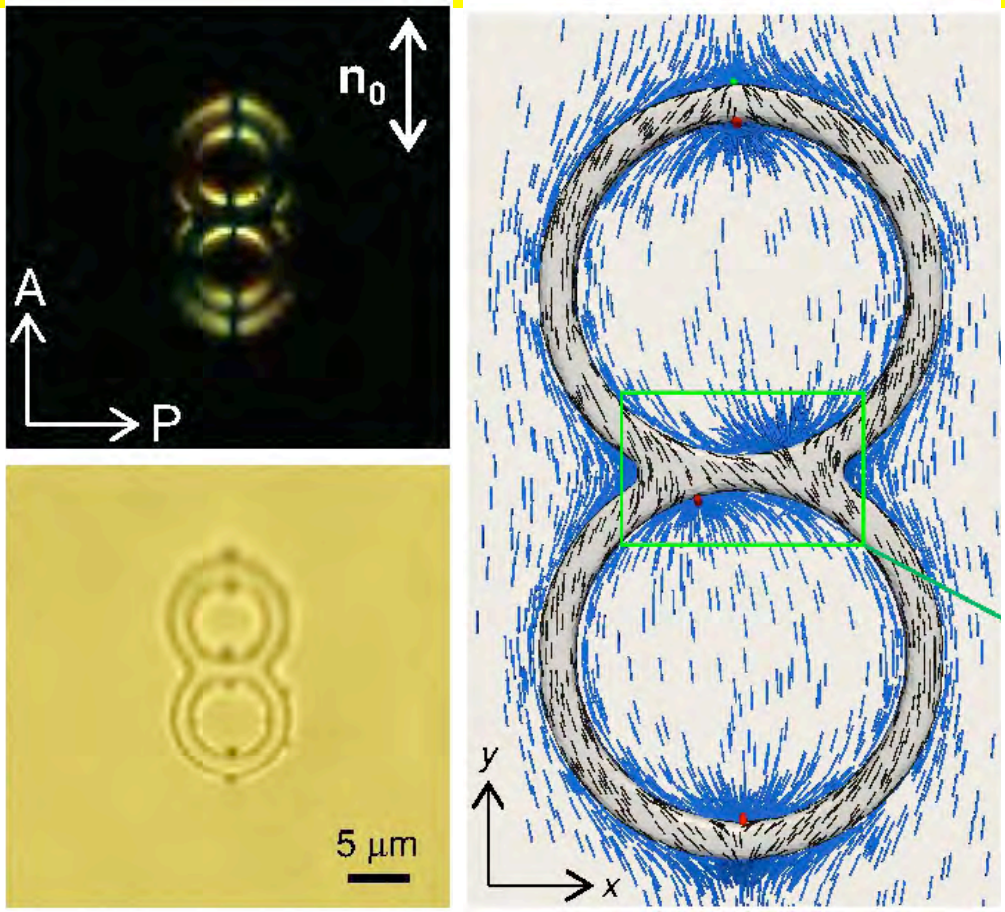




Energetic barrier - bistable

→Defects are not required but appear to reduce free energy!
→Still satisfy topological constraints (winding numbers add to zero)

Boojums on g=2 handlebodies

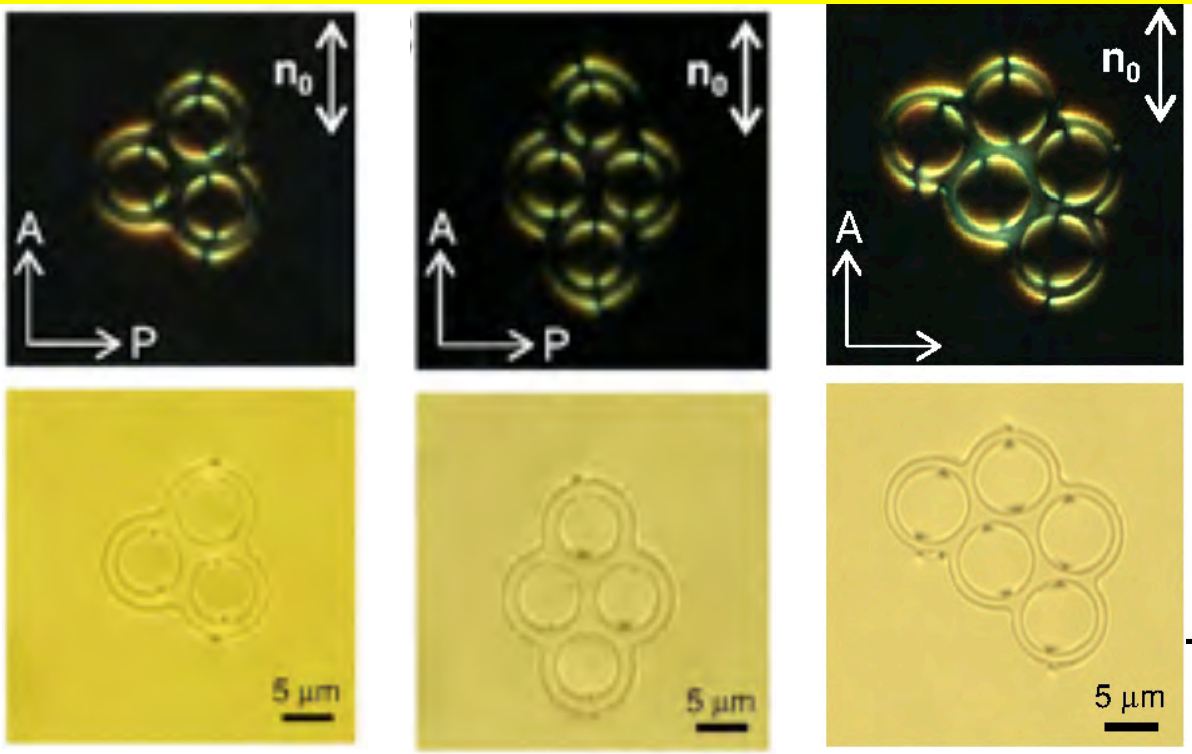


- **Different orientations, but the same net 2D winding numbers of boojums**
- **Satisfy predictions of topological theorems:**

$$\sum_i s_i = \chi = 2 - 2g$$

- **To induce boojums of net winding numbers *s* use a particle of right *g***

Boojums on colloidal handlebodies



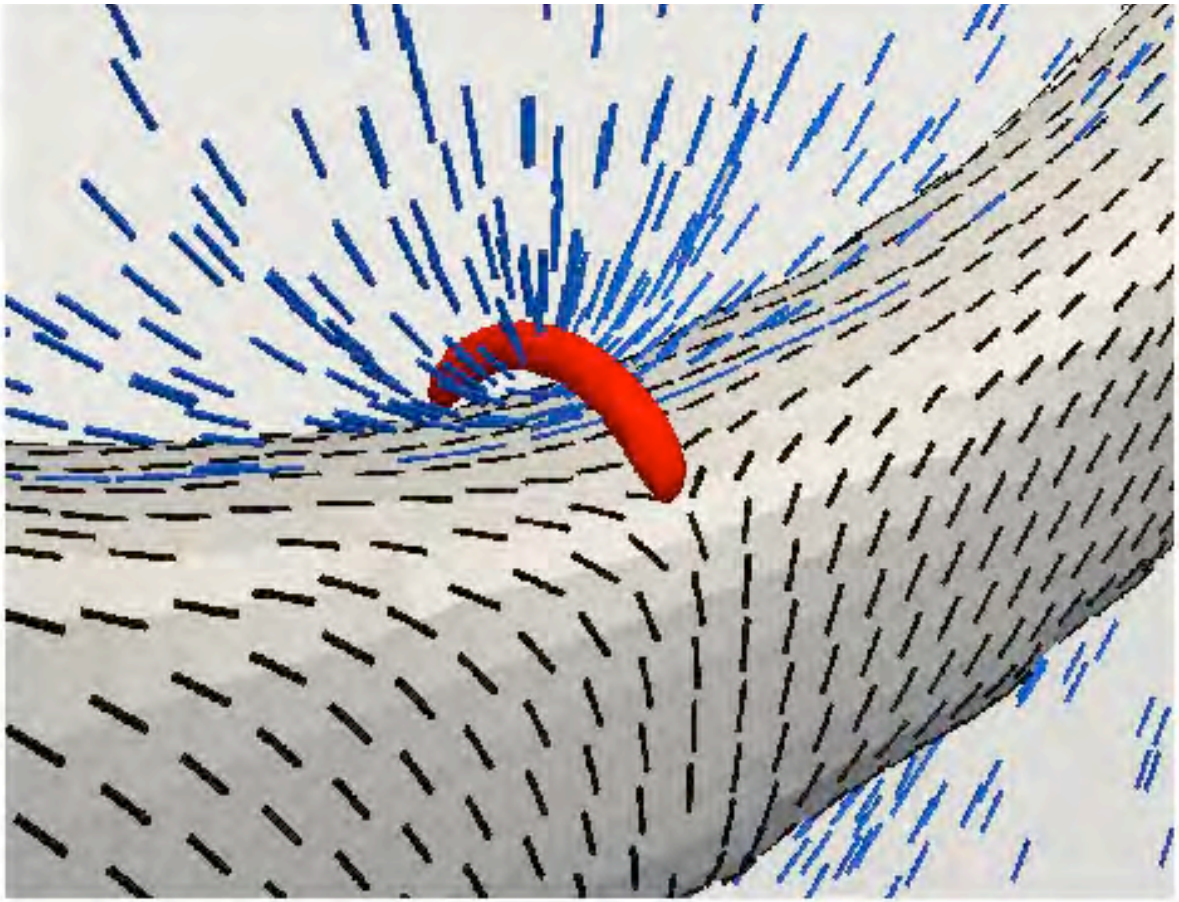
g=3,4,5 handlebodies

g	$\chi = \Sigma_i s_i$	Number of defects	
		s = +1	s = -1
1	0	2	2
		0	0
2	-2	2	4
		4	6
3	-4	2	6
		4	8
4	-6	2	8
		4	10
5	-8	2	10
		3	11

Theorems & charge conservation:

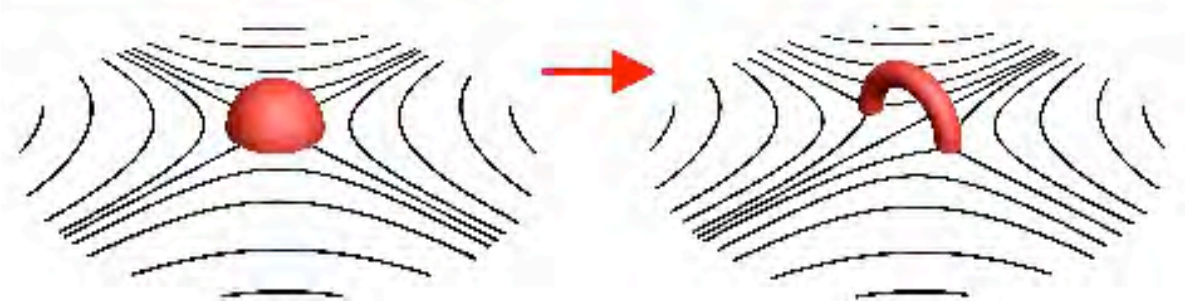
$$\sum_i s_i = \chi = 2 - 2g$$

Boojums **from handles** on colloidal handlebodies



Strong tangential anchoring promotes boojum splitting into disclination semirings

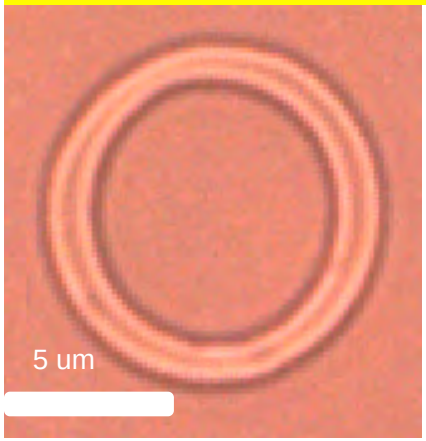
Splitting of boojum points to semi-rings increases *g:*
handle-shaped boojums on colloidal handlebodies

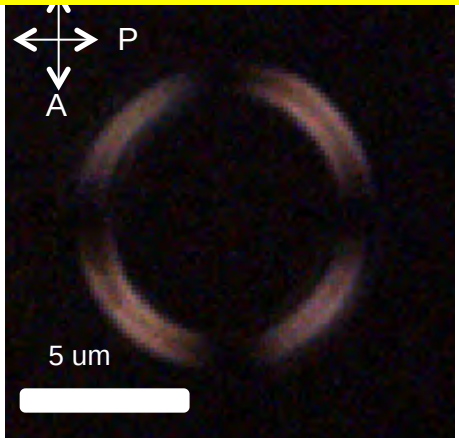


Q. Liu, B. Senyuk, M. Tasinkevych, and I. I. Smalyukh, Proc. Natl. Acad. Sci. U.S.A. **110**, 9231 (2013)

Drops of LCs with g>0?

Torus-shaped droplet confinement

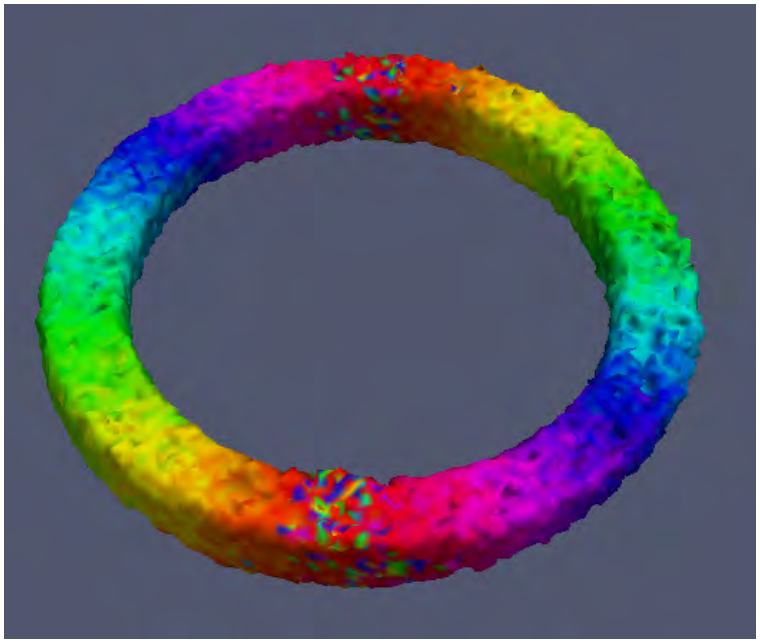


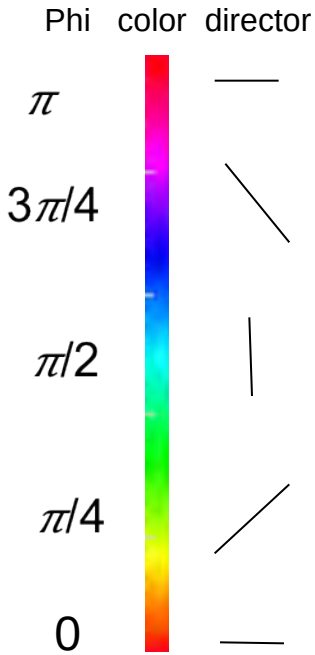


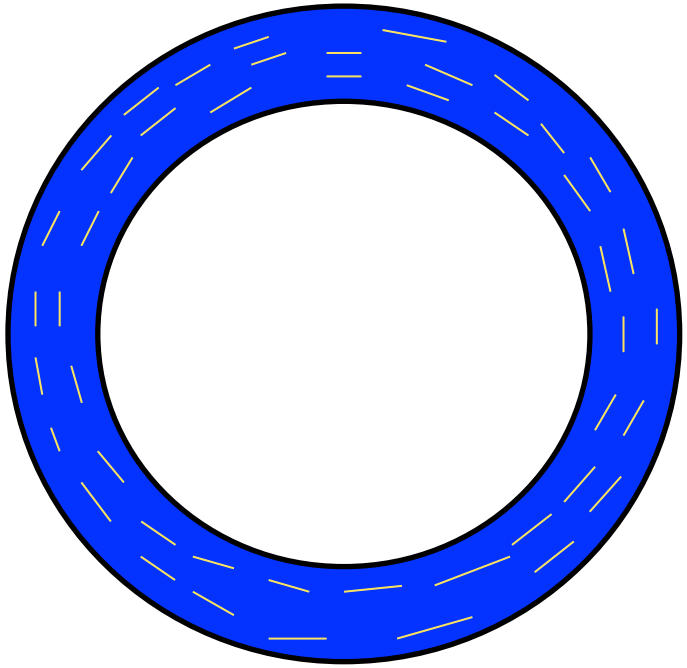
- No topological defects induced;

$$\sum_i s_i = \chi = 2 - 2g$$

- Director field reconstructed from 3D 3PEF-PM images

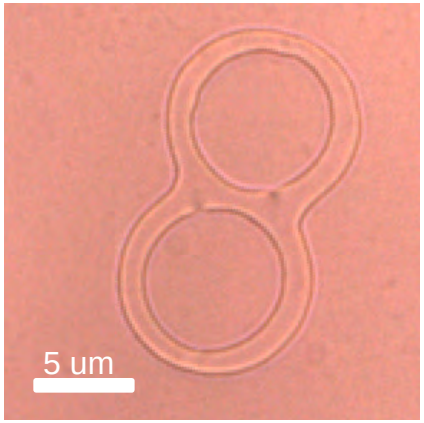


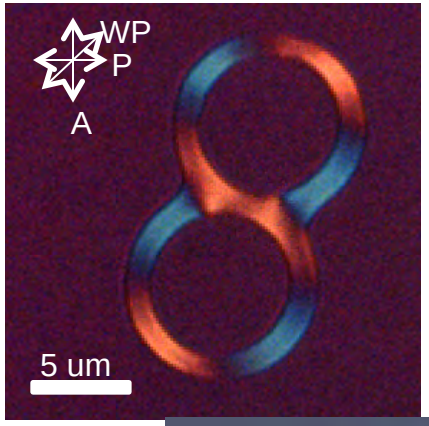


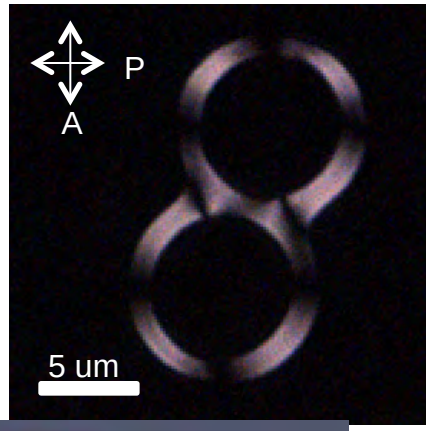


M. Campbell, M. Tasinkevych, I. Smalyukh. *Phys. Rev. Lett.* **112**, 197801 (2014).

g=2 Torus

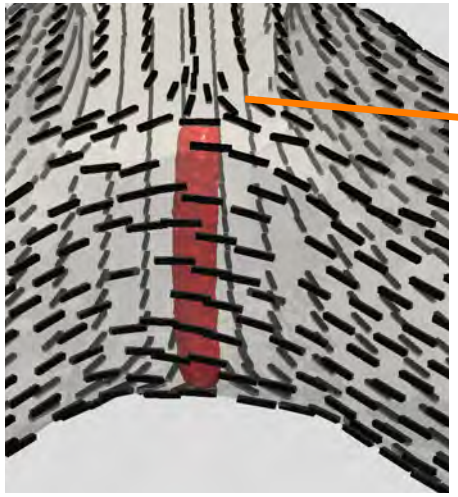


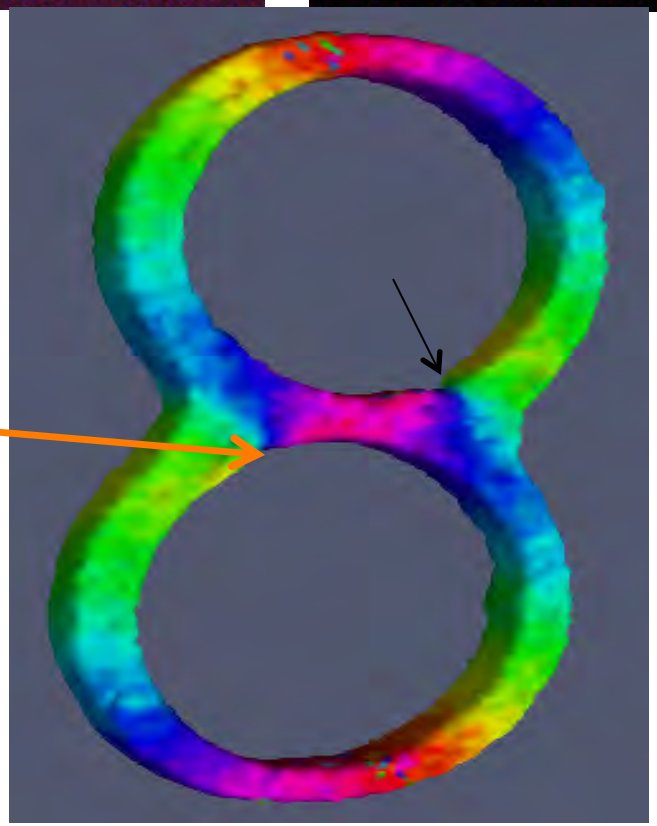


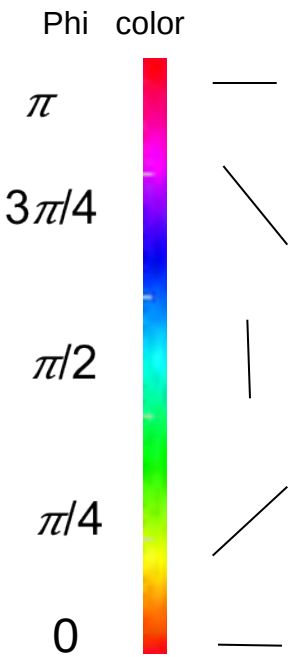


$$\sum_i s_i = \chi = 2 - 2g$$

s=-1/2 disclination

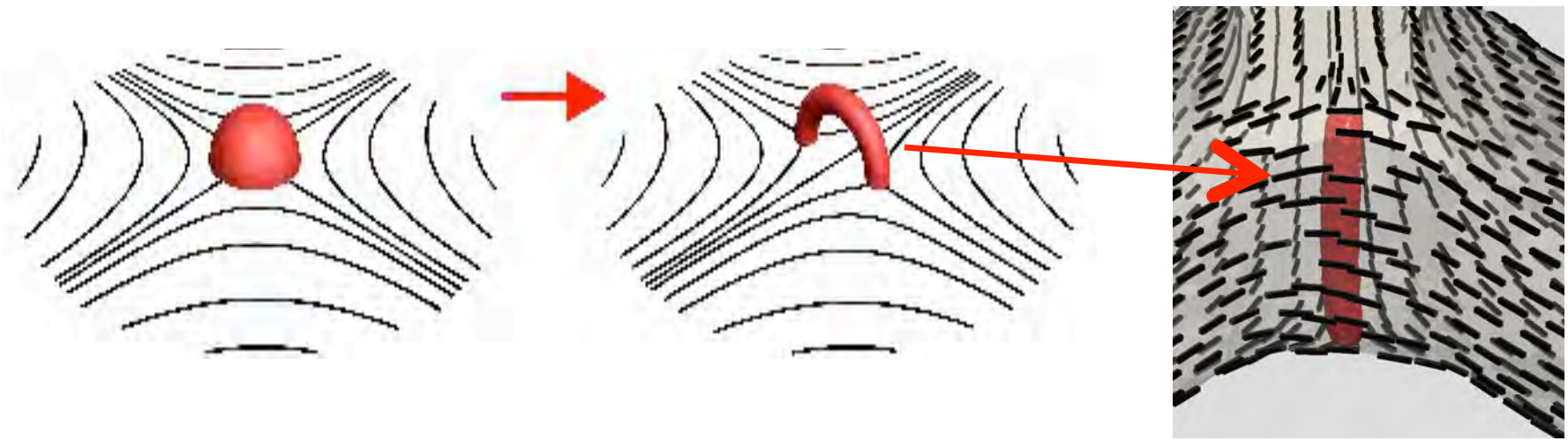






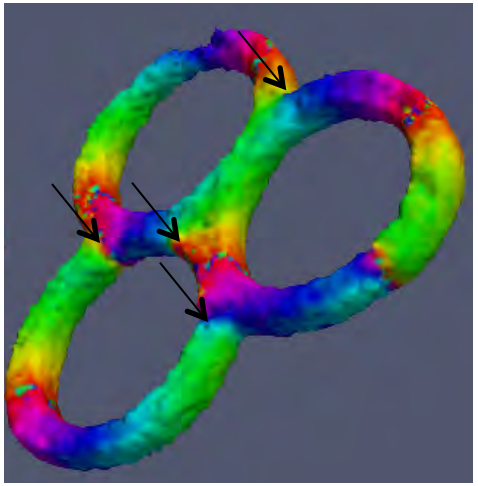
Defect Structure

- Rather than boojums, we observe line defects of winding number -1/2.
- These structures are topologically equivalent.
- The line defect is lower energy than the boojum in small drops.



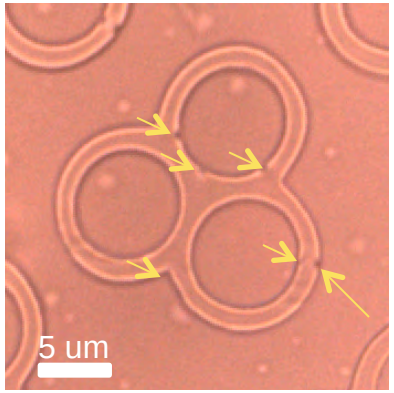
M. Campbell, M. Tasinkevych, I. Smalyukh. *Phys. Rev. Lett.* **112**, 197801 (2014).

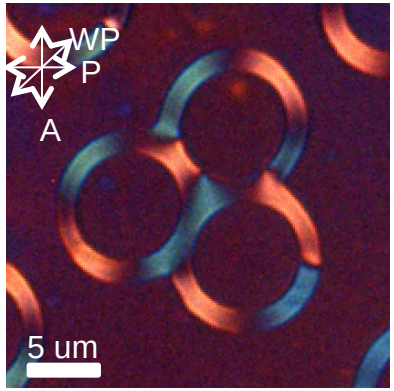
g=3 torus

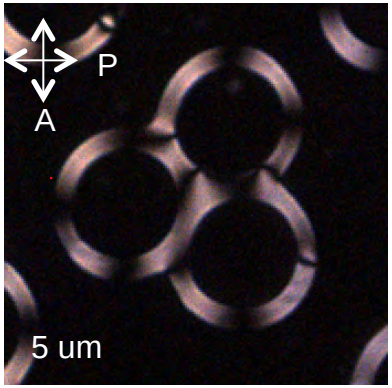


- The defects are -1/2 line defects spanning across the pore thickness
- They are located at the intersection of the three tori
- The strength at surfaces adds to satisfy topology

$$\sum_i s_i = \chi = 2 - 2g$$



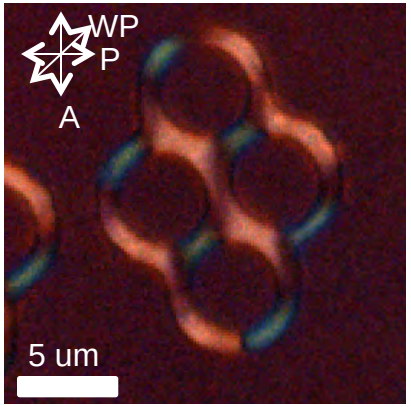


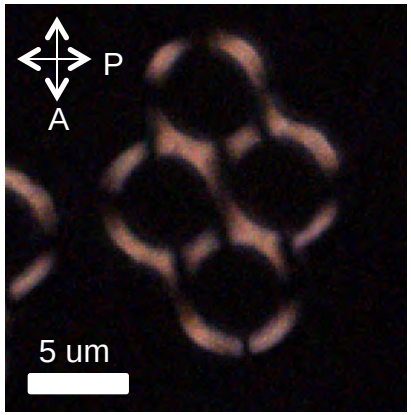


M. Campbell, M. Tasinkevych, I. Smalyukh. *Phys. Rev. Lett.* **112**, 197801 (2014).

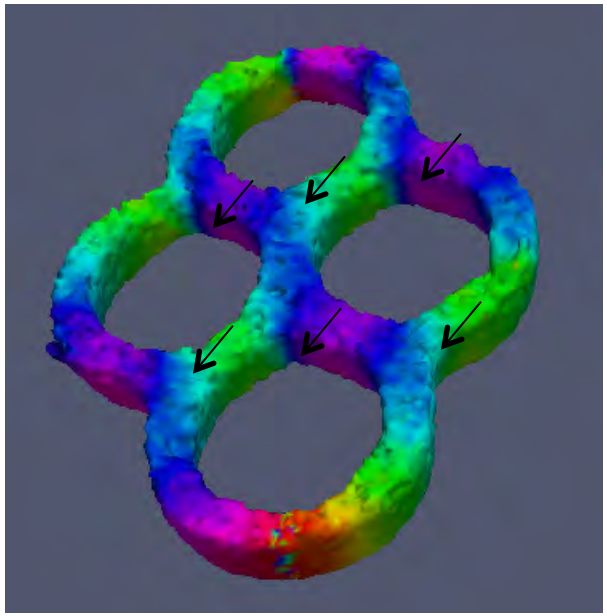
g=4 and g=5 tori

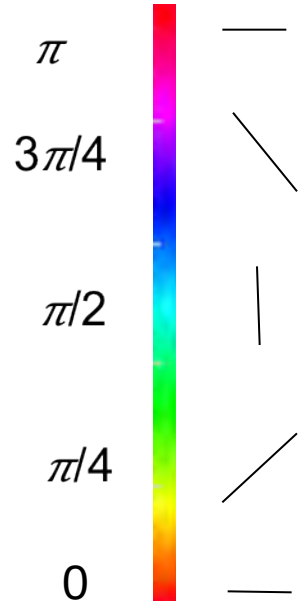
minimum 6 defects



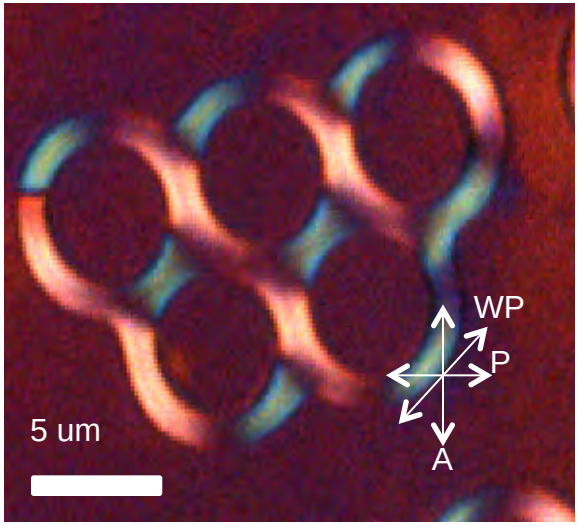


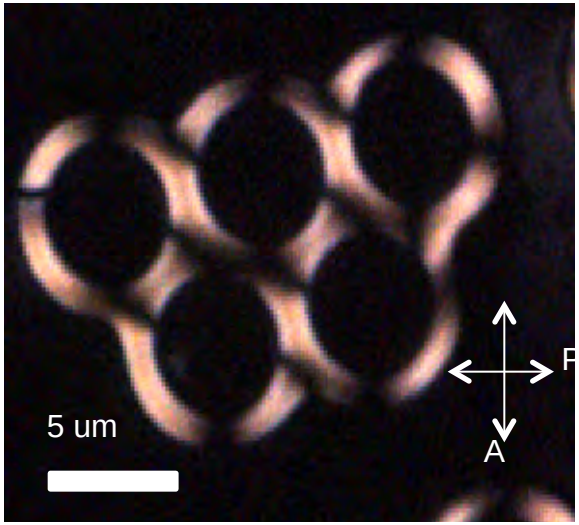
$$\sum_i s_i = \chi = 2 - 2g$$

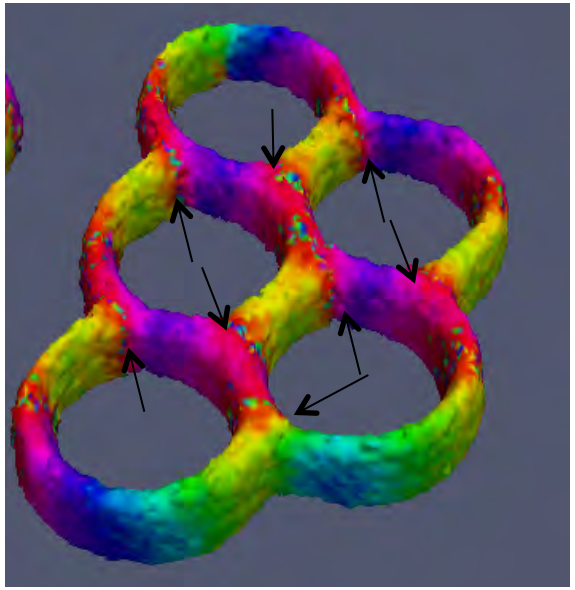




minimum 8 defects

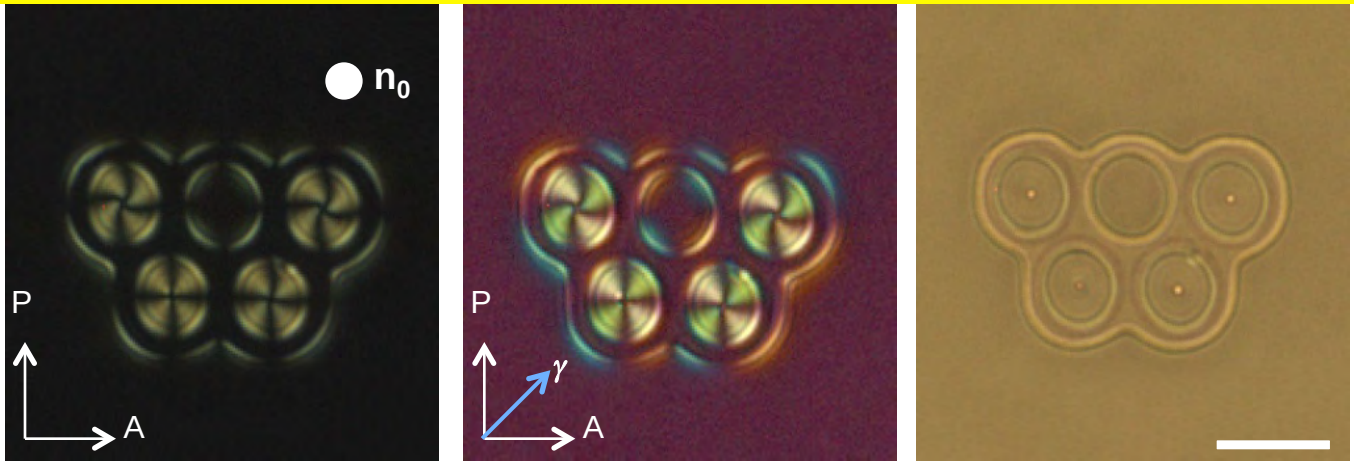




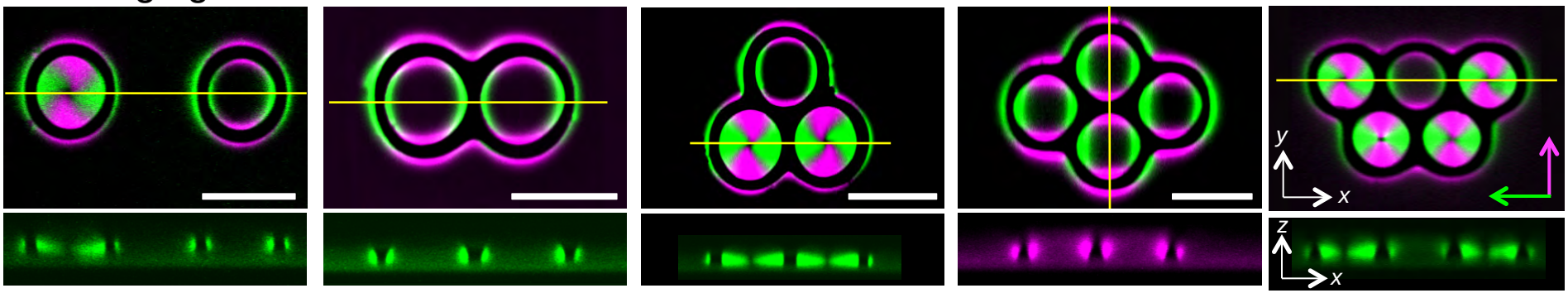


Handlebody-shaped colloids, perpendicular BCs

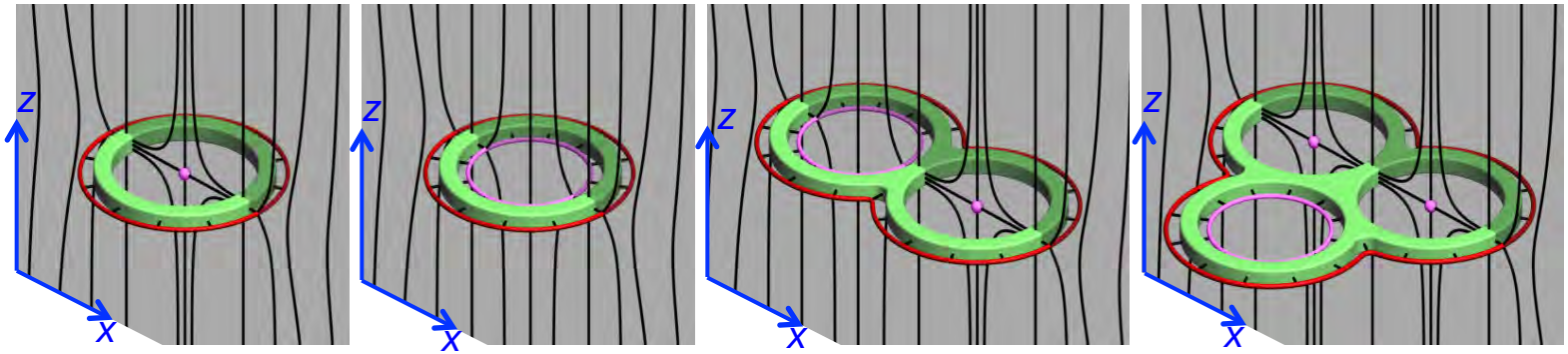
Polarizing optical m-py



3D imaging



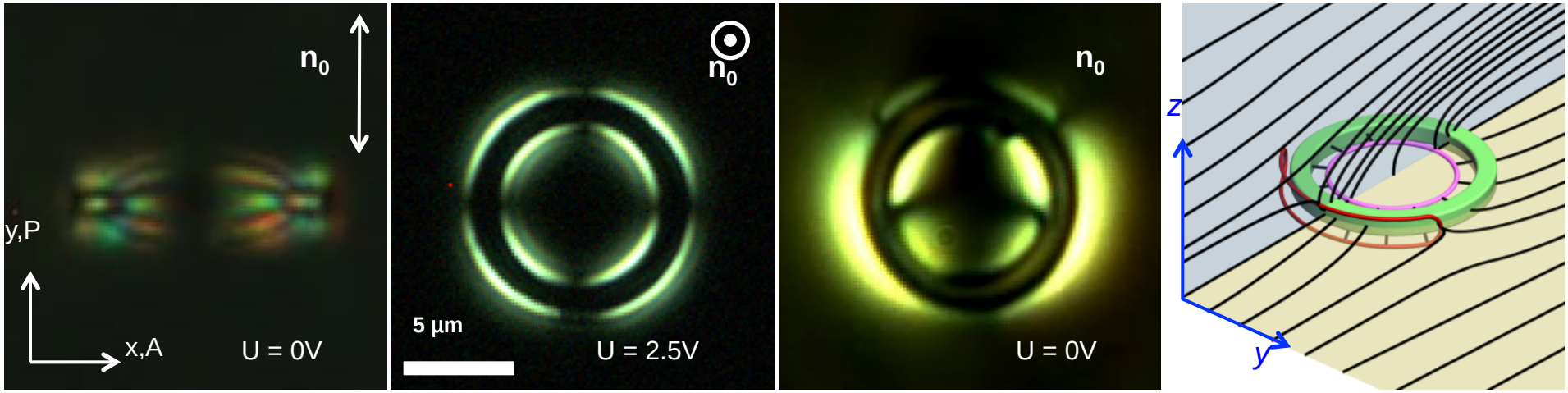
Reconstructed director structures and defects around colloidal hanlebodies

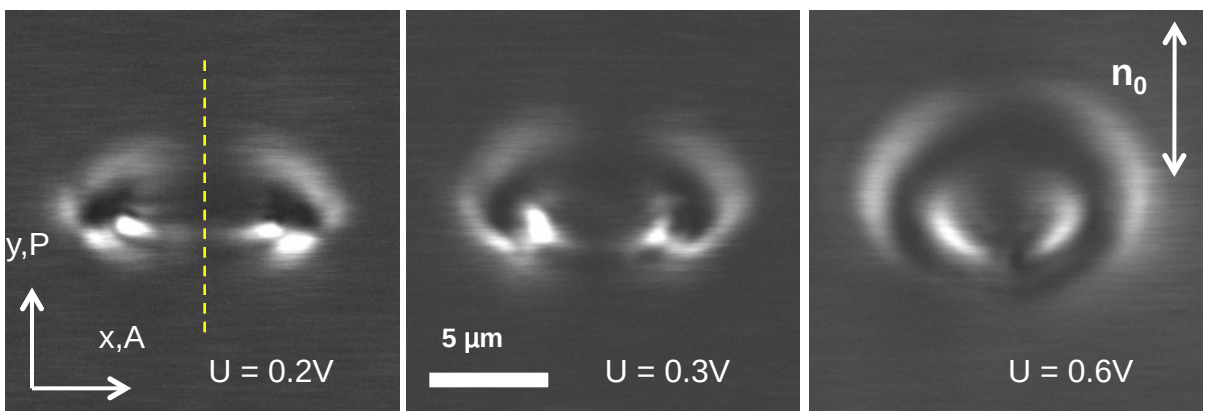


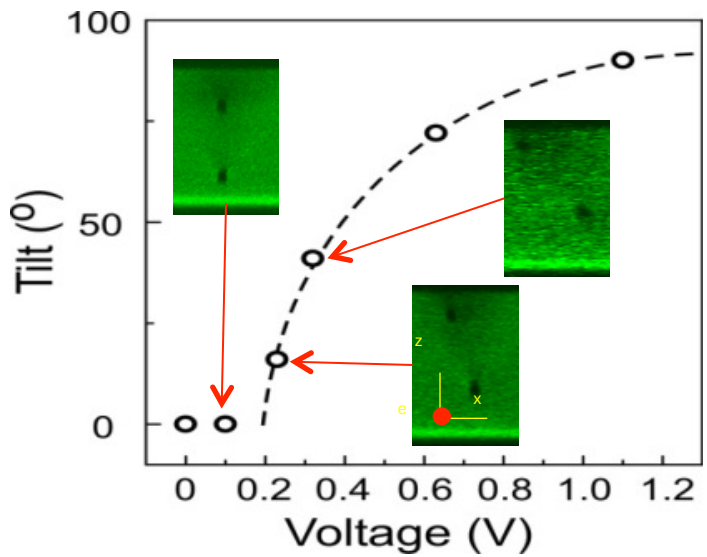
Senyuk, Liu, He, Kamien, Kusner, Lubensky, & Smalyukh, *Nature* **493**, 200-205 (2013)

Switching director & rotating colloidal tori

- **Transformation of defects in applied electric field;**
- **Bistable particle orientations & fields at no field;**



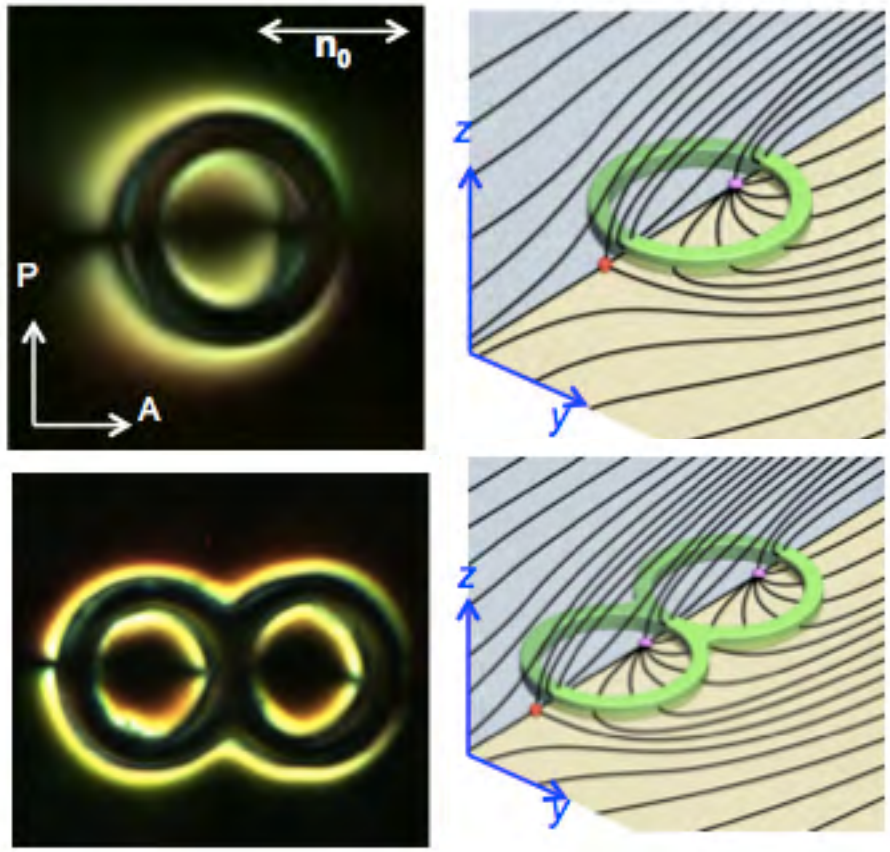




B. Senyuk, Q. Liu, S. He, R. Kamien, R. Kusner, T. Lubensky, & I.I. Smalyukh, " Nature **493**, 200-205 (2013)

Transforming LC defects around colloidal tori

Alignment parallel to the far-field director



Nature **493**, 200 (2013)

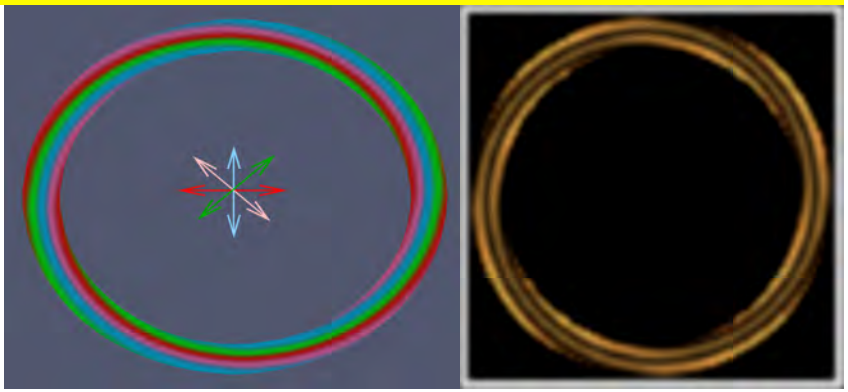
- **In all configurations bulk defects compensate the charge of particle;**
- **Satisfy predictions of the Poincare-Hopf index theorem:**

$$\sum_i m_i = \pm\chi / 2 \qquad \chi = 2 - 2g$$

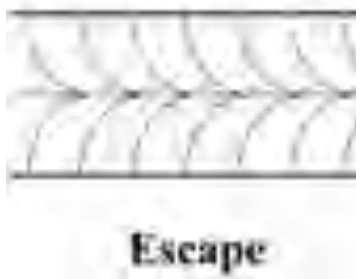
- **Induce net topological charge m (mod 2) by a particle of right g**

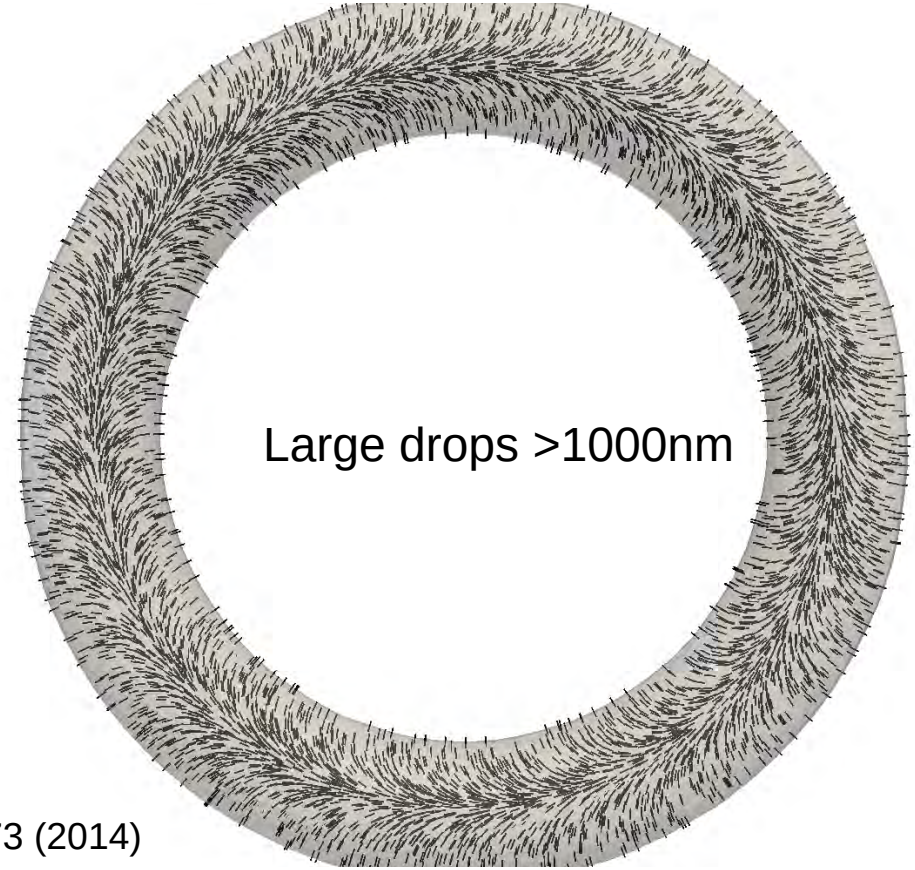
Perpendicular BCs: g=1 torus

- Escape in 3D dimension:
  - in cylindrical capillary;
  - under torus confinement;



$$\sum_i m_i = \pm\chi / 2$$

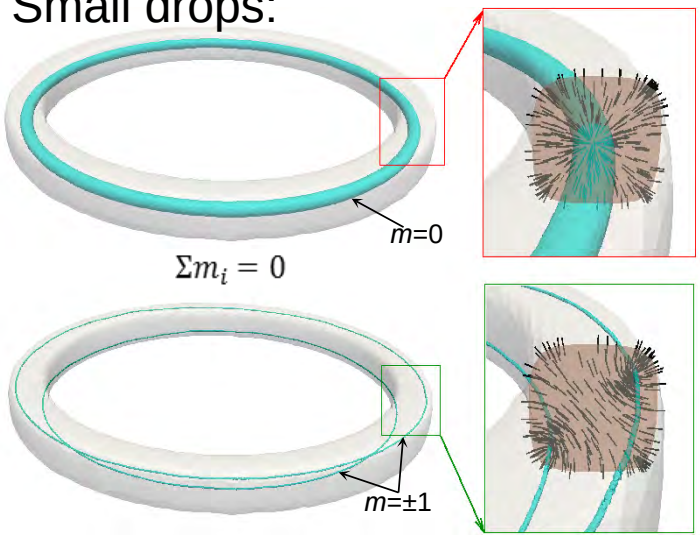




Tasinkevych, Campbell, Smalyukh, PNAS. **111**, 16268-16273 (2014)

g=1 confinement with knotted/linked defect lines

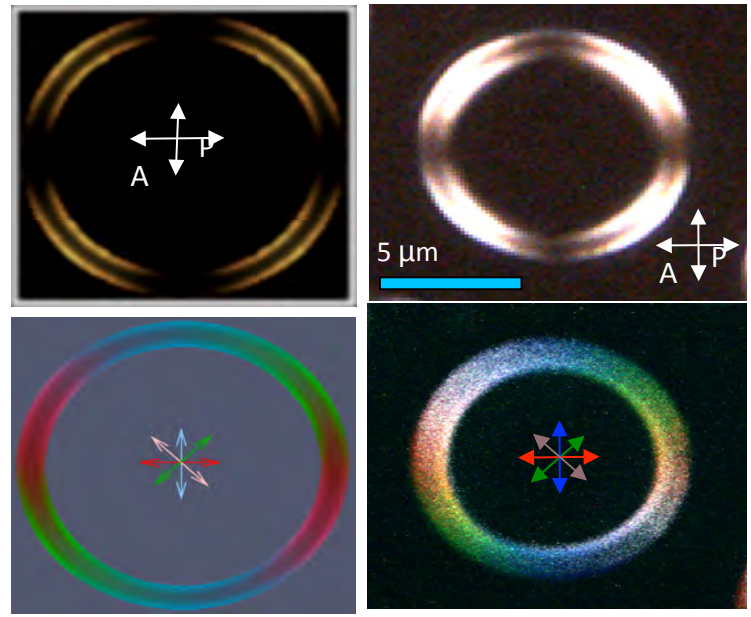
Small drops:



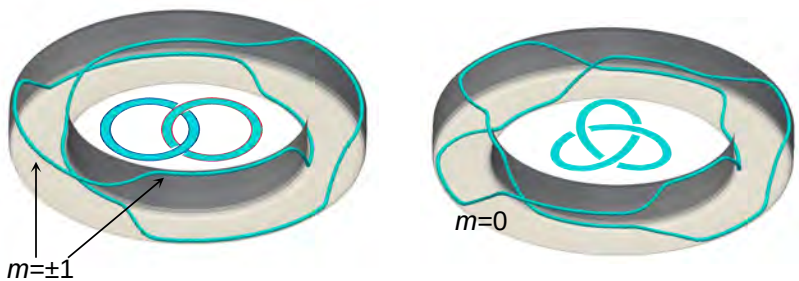
15-50 nm

50-1000 nm

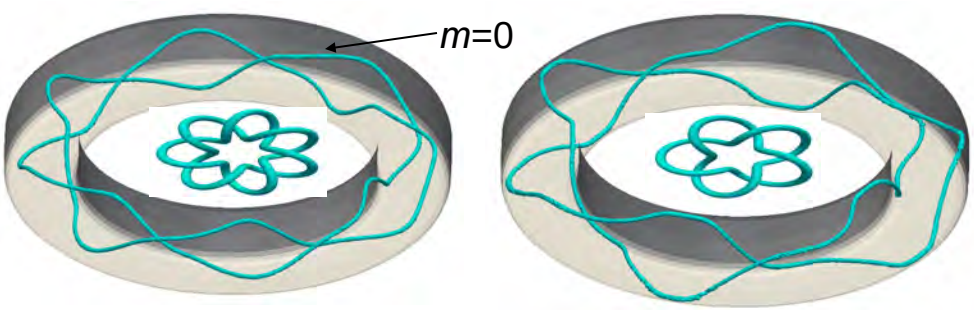
Image comparison:

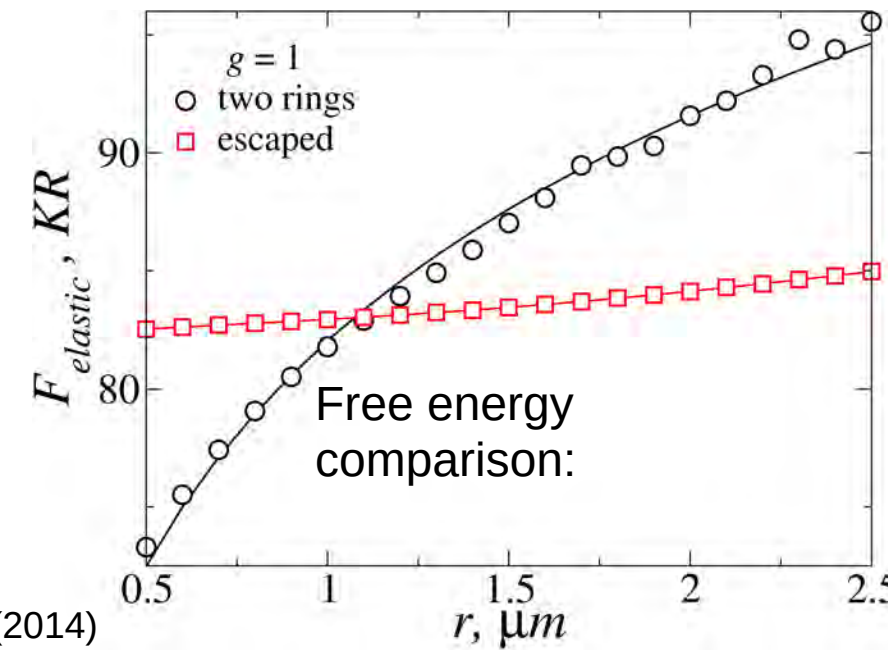


Links & trefoil knots:



Other torus knots:



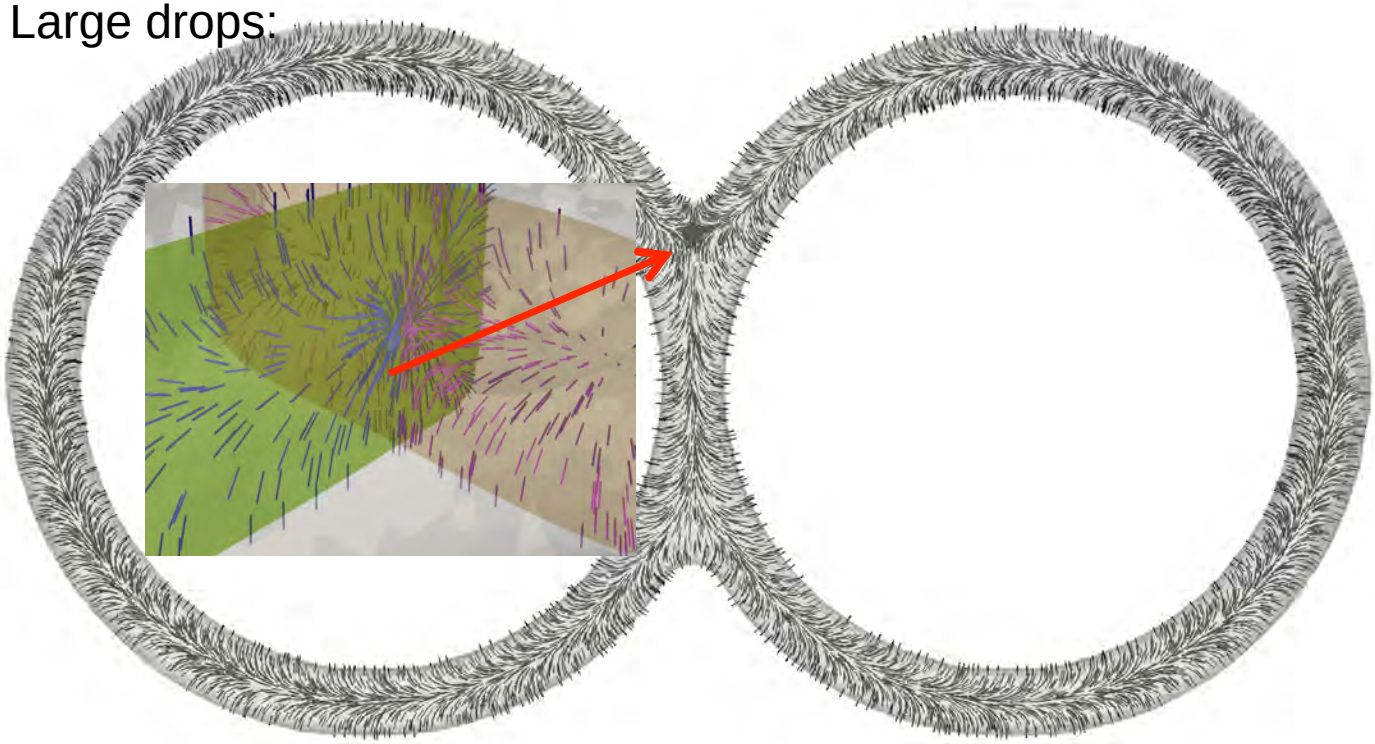


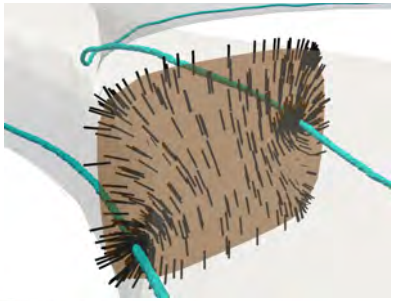
Free energy comparison:

Tasinkevych, Campbell, Smalyukh, PNAS. **111**, 16268-16273 (2014)

Equilibrium structures in g=2 drops

Large drops:





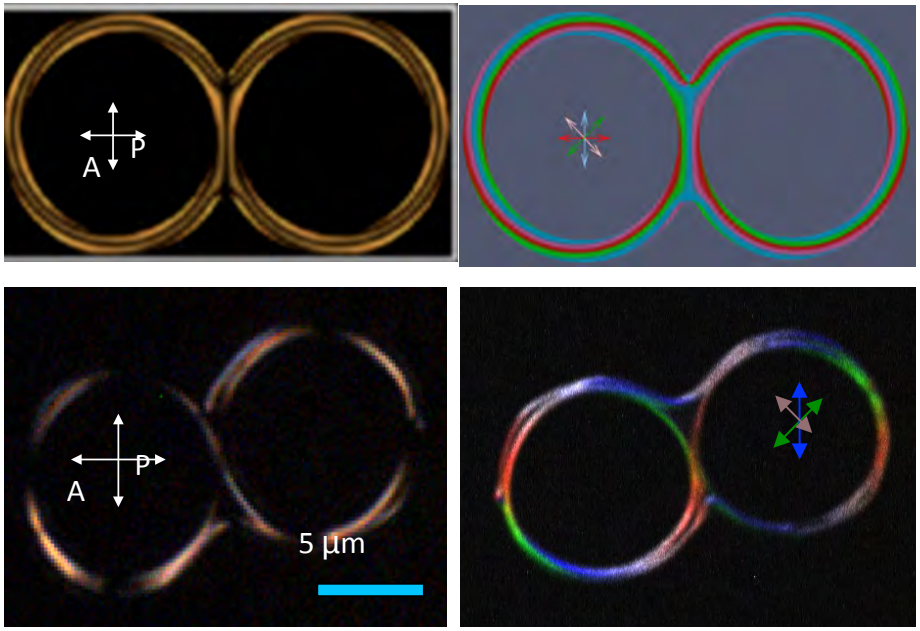
Small drops:

$$\sum_i m_i = \pm\chi / 2$$

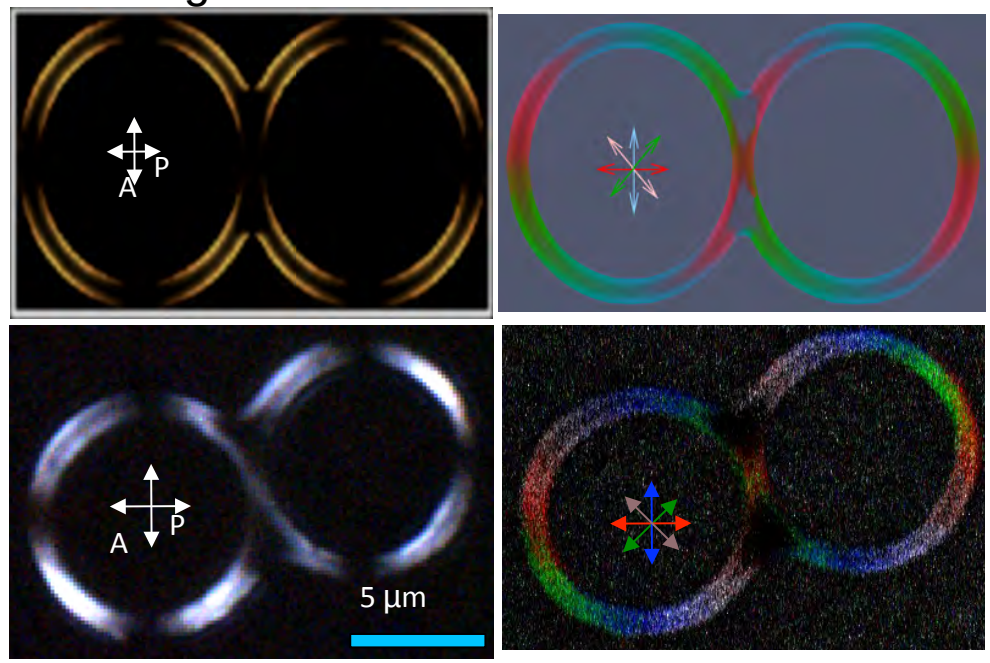
M. Tasinkevych, M. Campbell, I. Smalyukh, PNAS. 111, 16268-16273 (2014)

Imaging and Elastic Energy

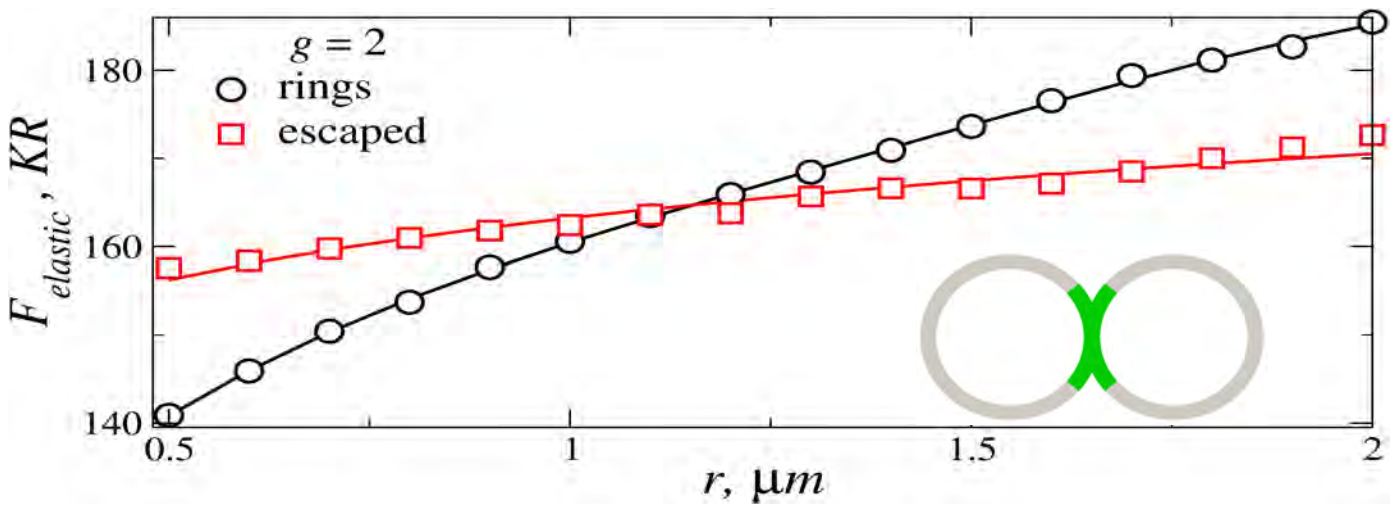
Escaped:



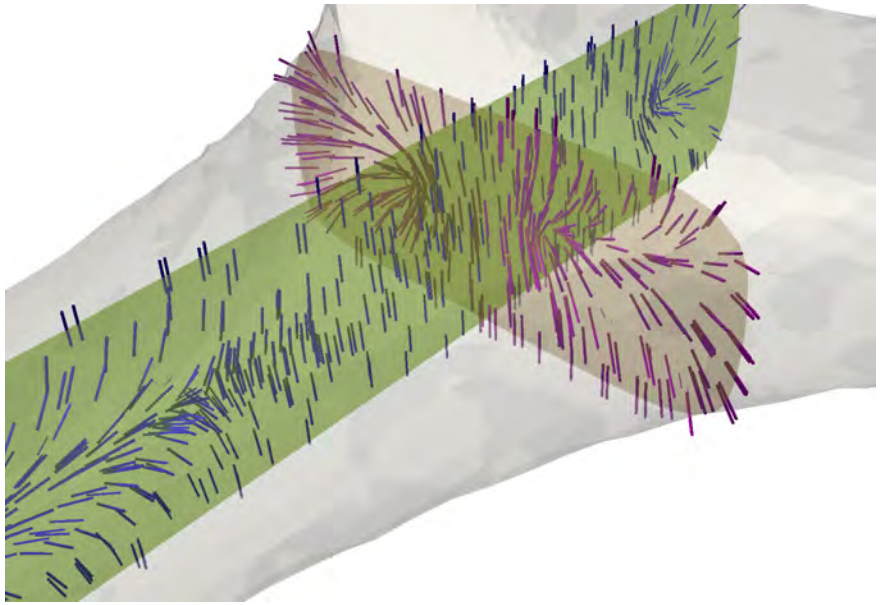
With singular defect lines:



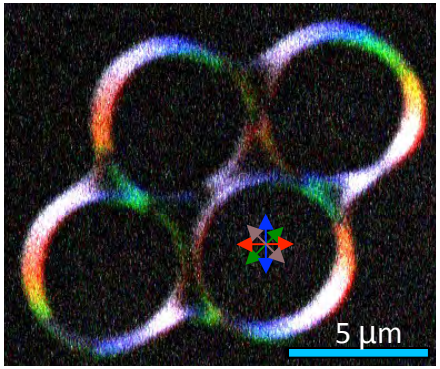
Free energy comparison:

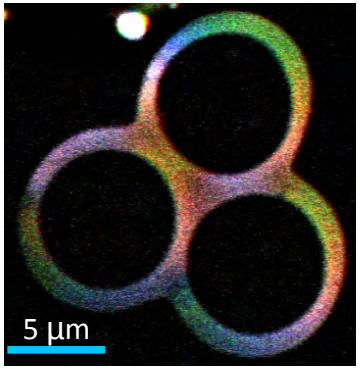


g=4,5 handlebody confinement with "escaped" director field

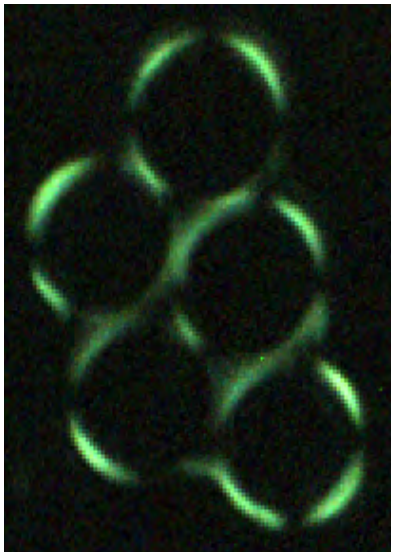


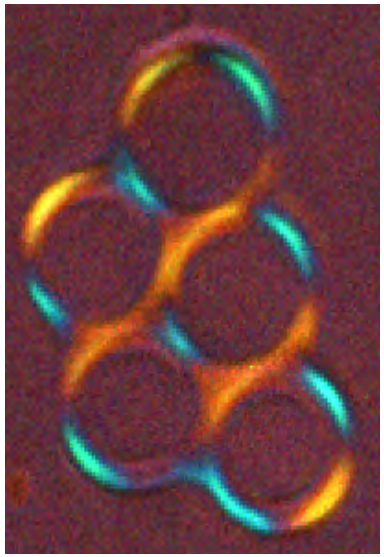
Nonlinear optical images:

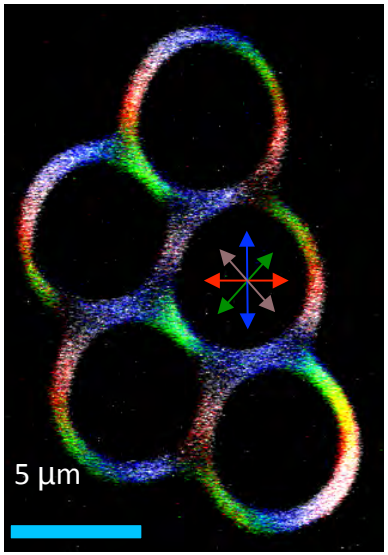




Polarizing optical m-py:

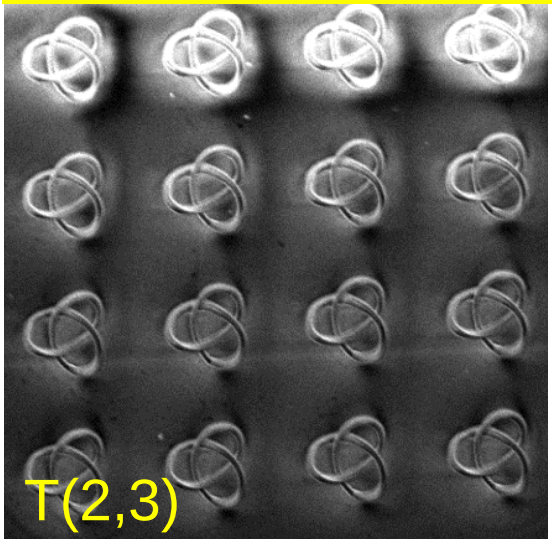


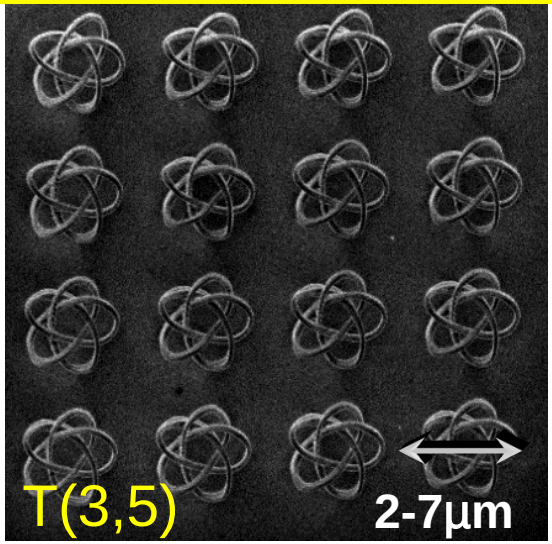


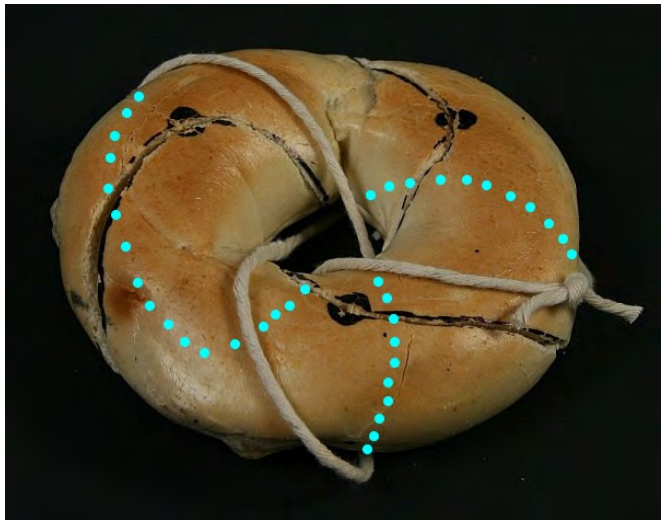


$$\sum_i m_i = \pm\chi / 2$$

Knot-shaped topological colloids (SEM)

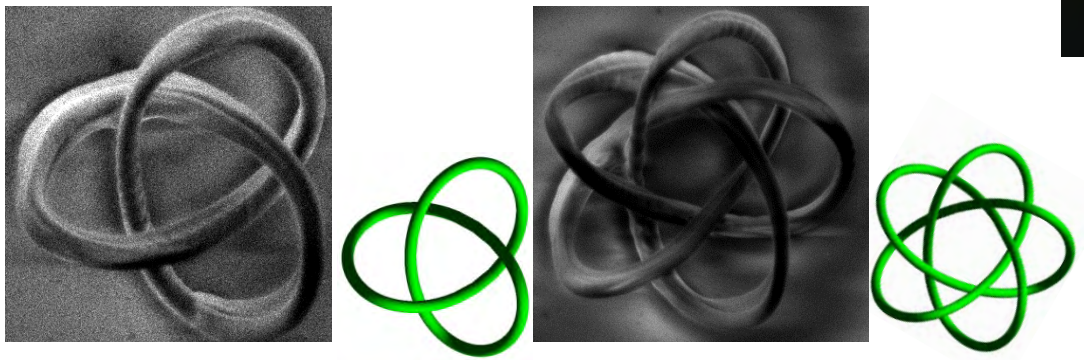




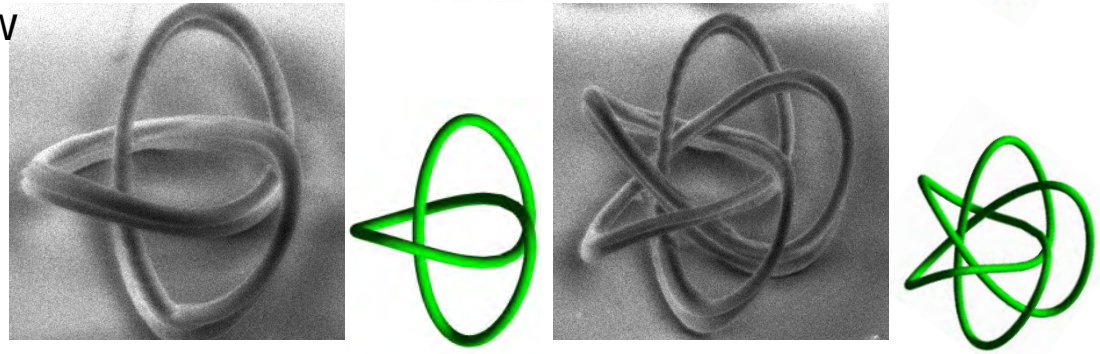


http://georgehart.com/bagel/knot.html

top view



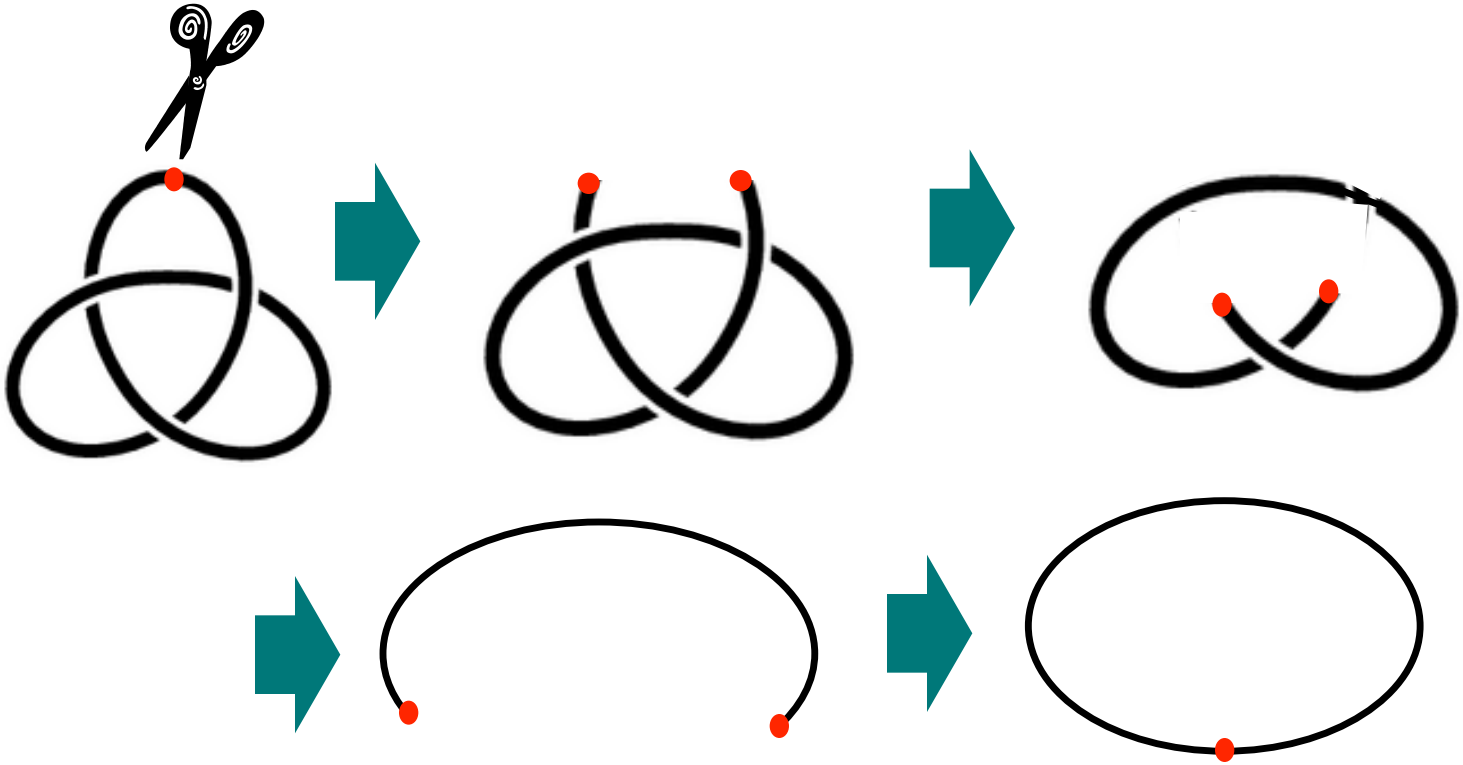
side view



What defects can be induced by knotted particles?

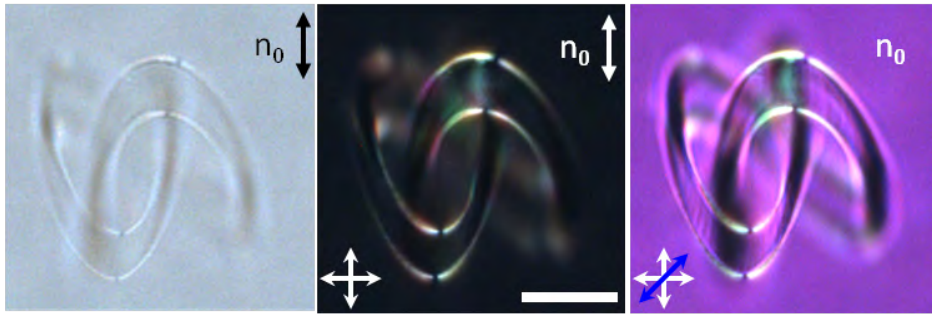
T(P,Q) knot winds Q times around a circle in interior of a torus & P times around its axis

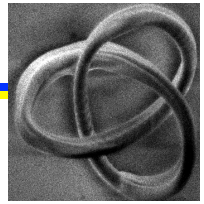
Euler characteristic of a torus knot



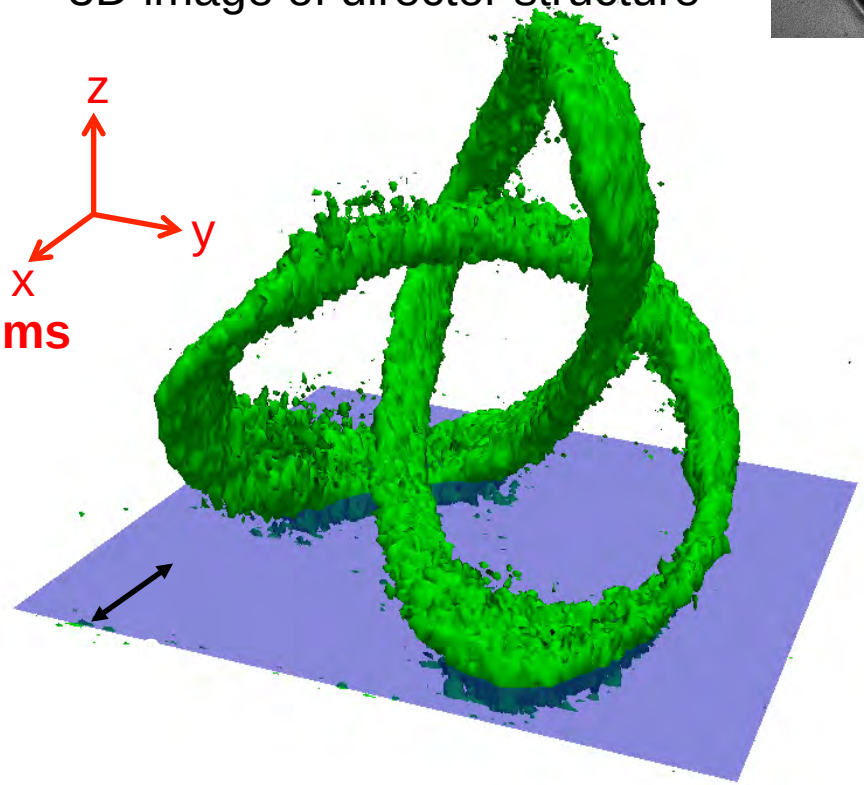
→**Euler characteristic of torus knots is χ=0**

Trefoil Colloidal-knot-induced defects

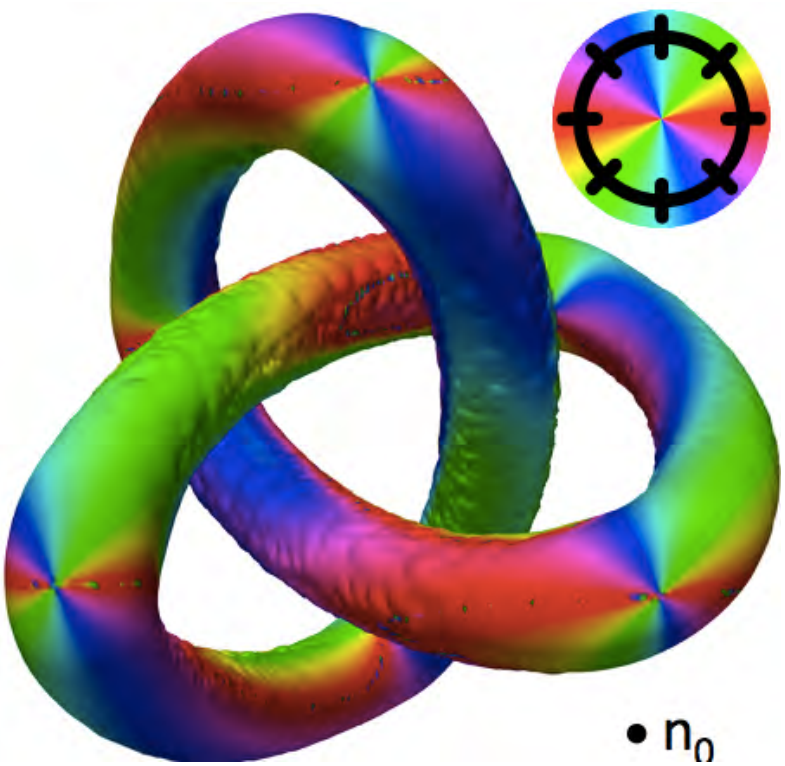




3D image of director structure



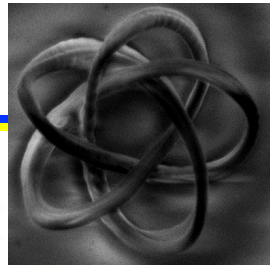
Knots induce 12 self-compensating boojums

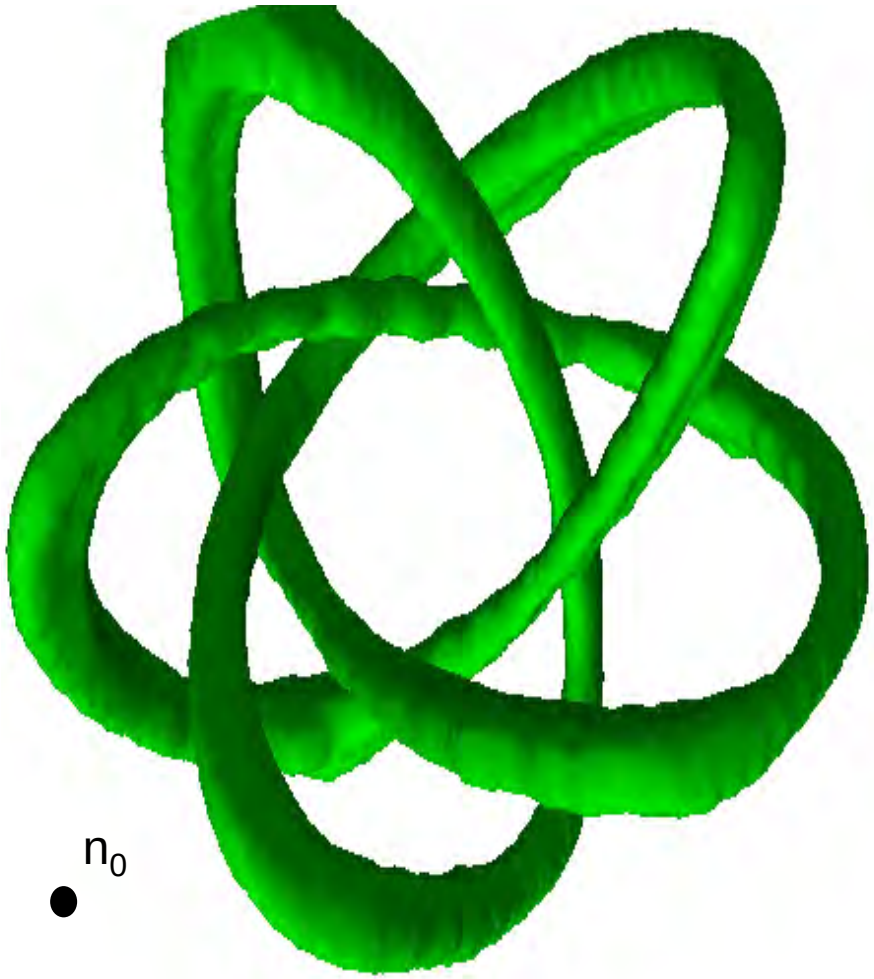


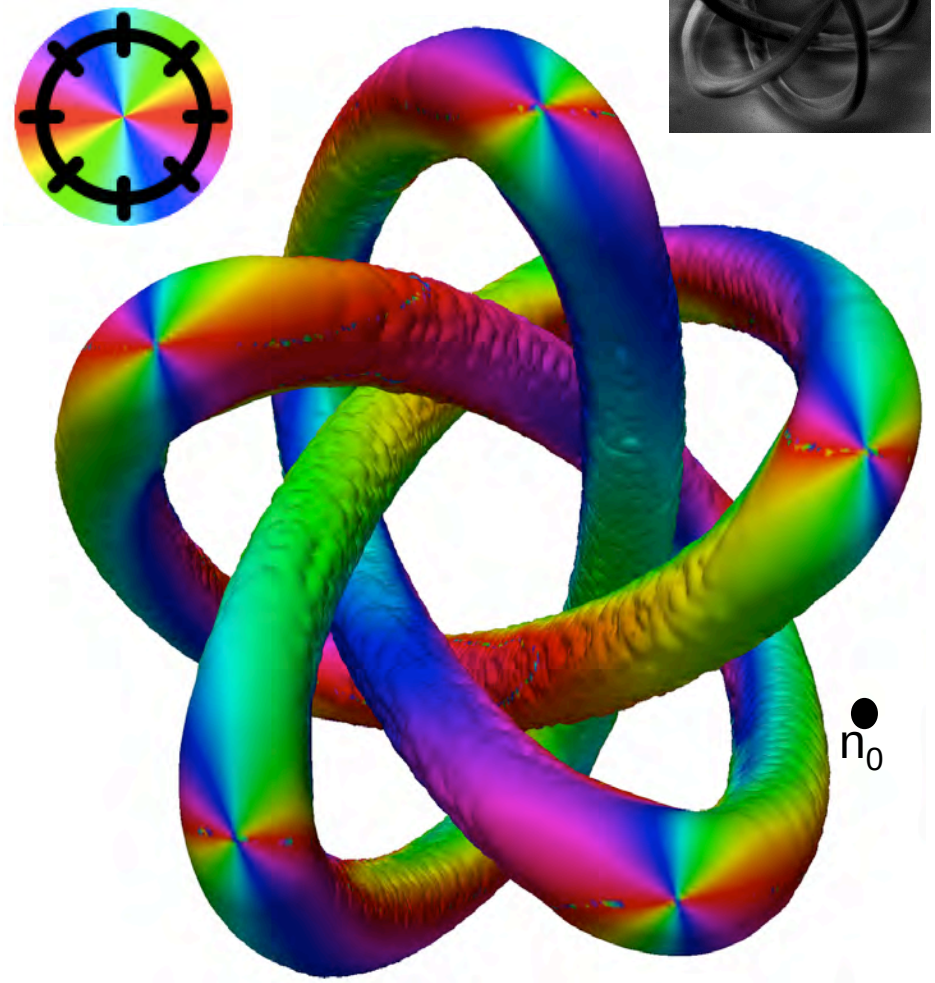
→6 *s*=1 & 6 *s*=-1 boojums compensate each other
→ consistent with χ=0
→T(p,q)=T(2,3)
→The total number of boojums is 4q

A. Martinez, M. Ravnik, B. Lucero, R. Vishvanathan, S. Zumer, & I.I. Smalyukh, *Nature Mater* (2014)

T(3,5) colloidal-knot-induced defects







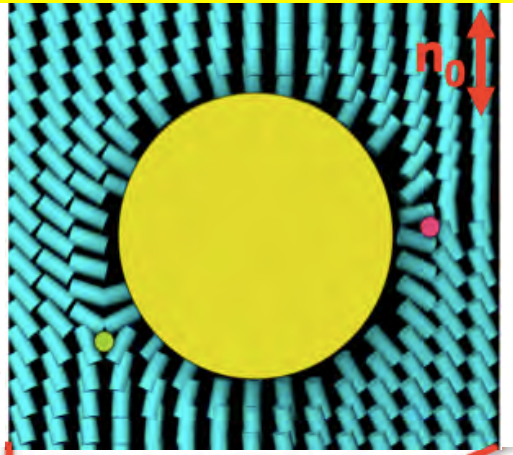
→10 *s*=1 & 10 *s*=-1 boojums compensate each other

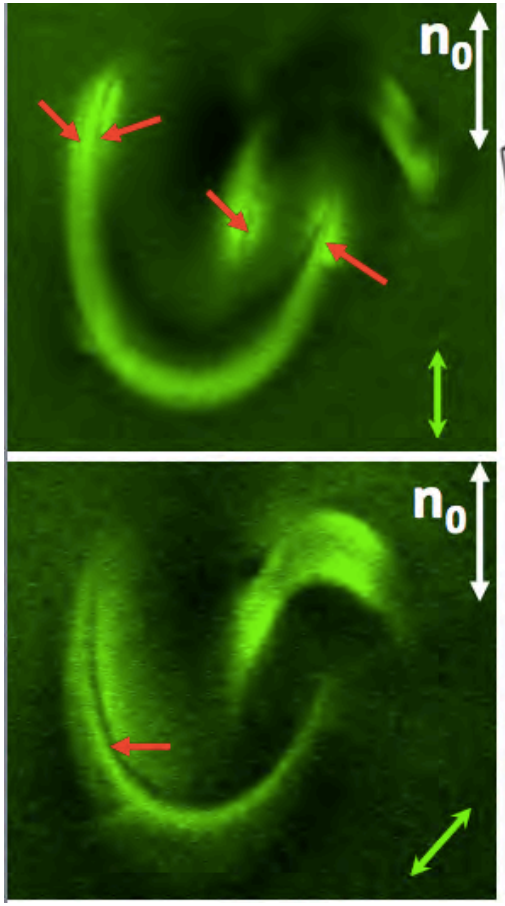
→ consistent with χ=0

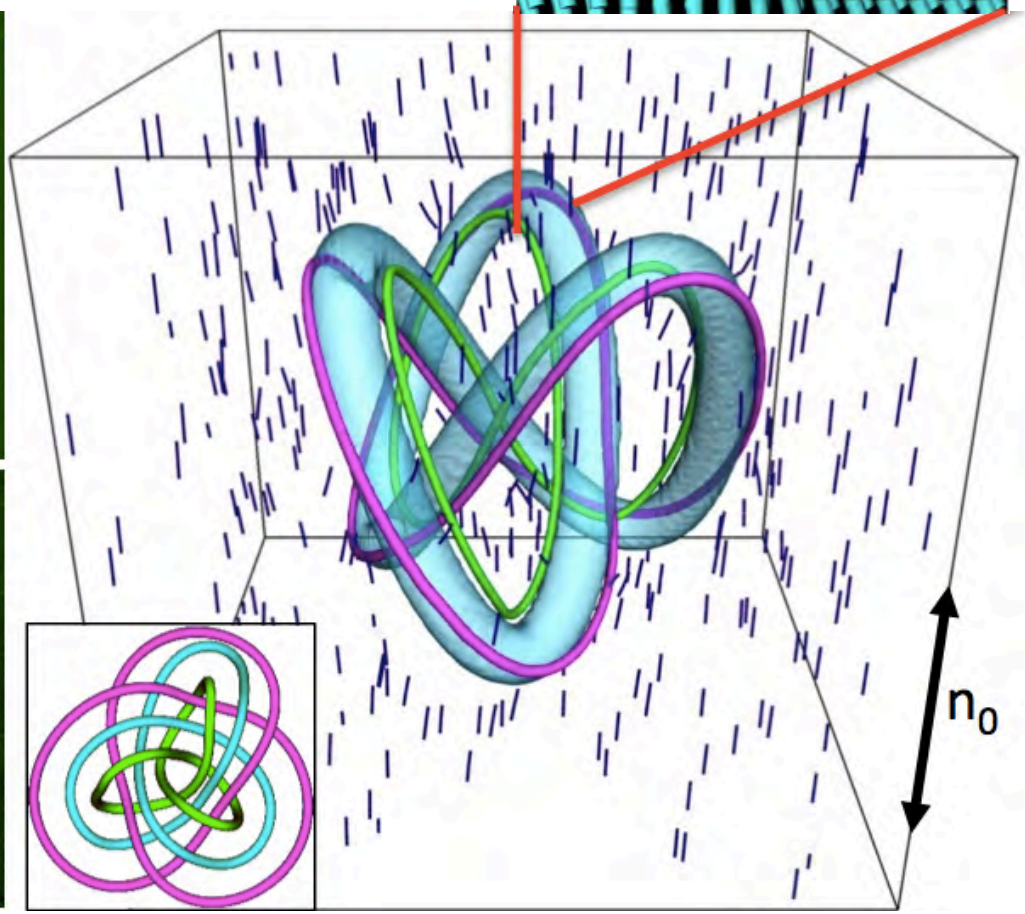
→The total number of boojums is 4q again

Mutually tangled particle knots & director field

→Trefoil knot colloidal particle;
→perpendicular BCs;
→two knotted s=-1/2 defect lines;
→Mutually tangled particle and defect lines!
→Insights into integrations of knotted fields & surfaces

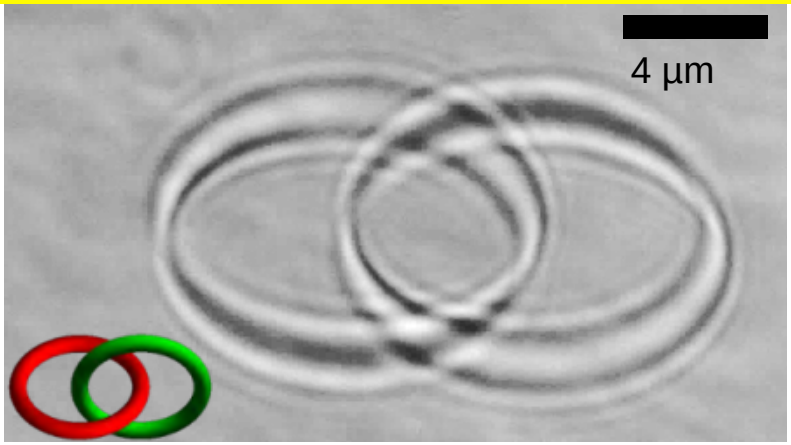




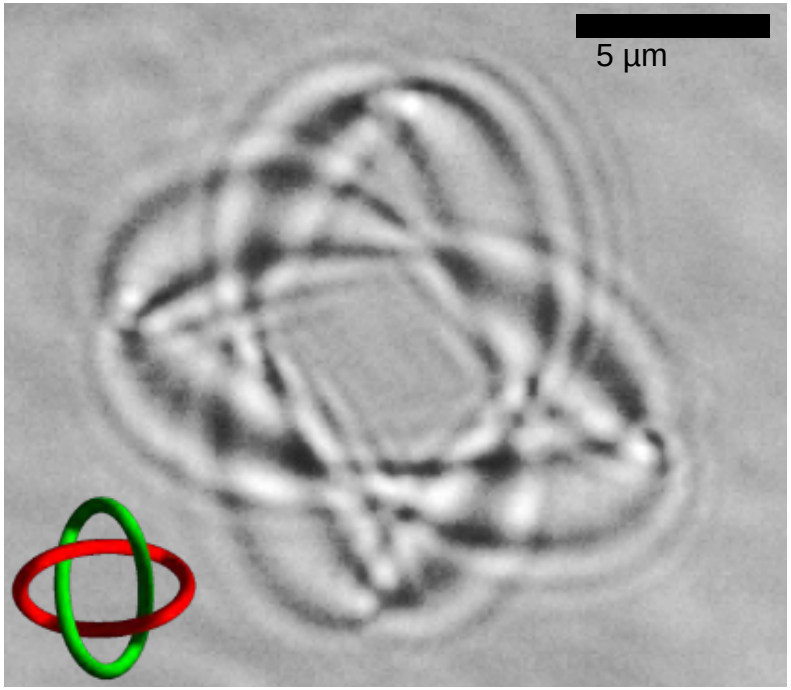


Multi-component particles lacking surface connectivity: colloidal links

Hopf Links

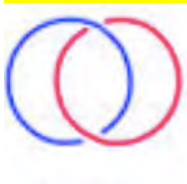


Solomon Links

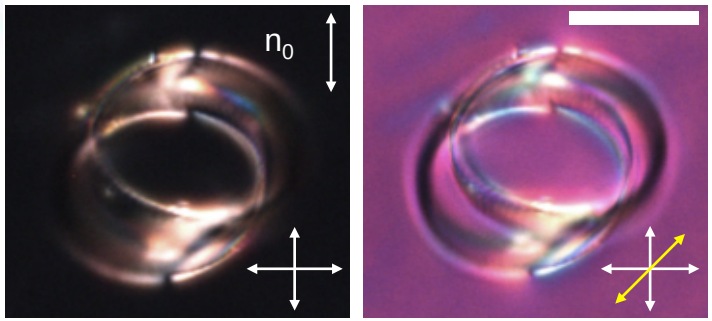


A. Martinez, L. Hermosillo, M. Tasinkevych, and I. I. Smalyukh. PROC. NATL. ACAD. SCI. U.S.A. 112, 4546-4551 (2015)

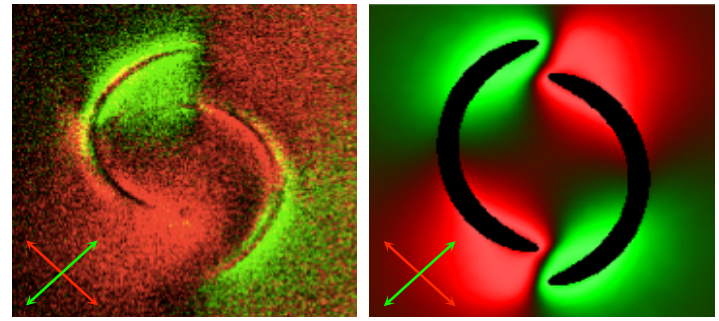
Colloidal Hopf Links with tangential BCs



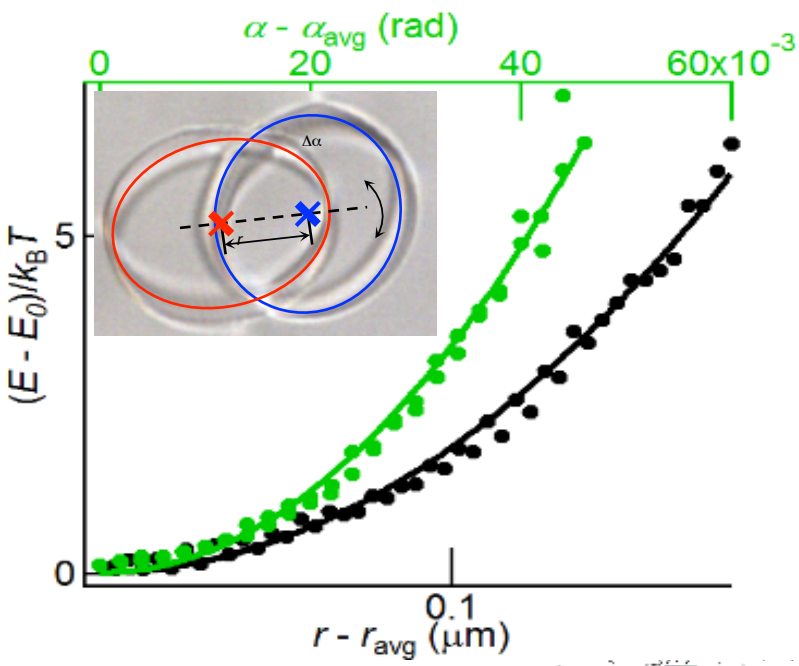
Polarizing optical microscopy

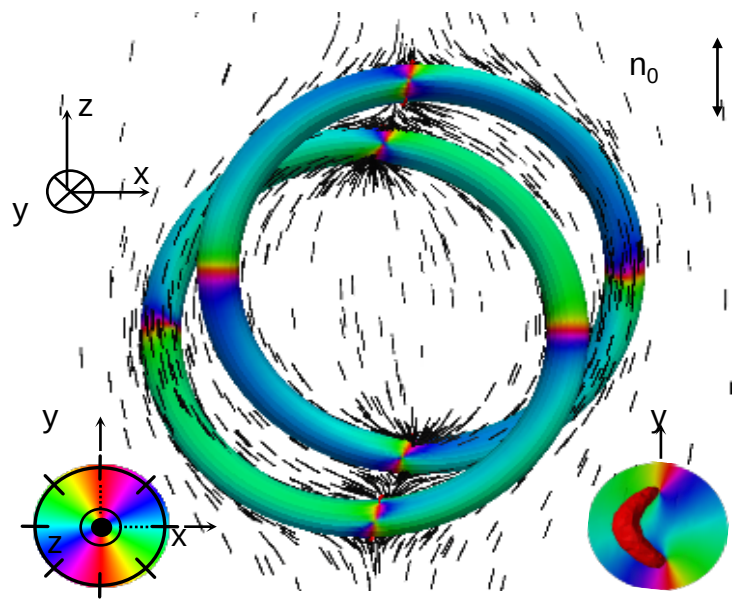


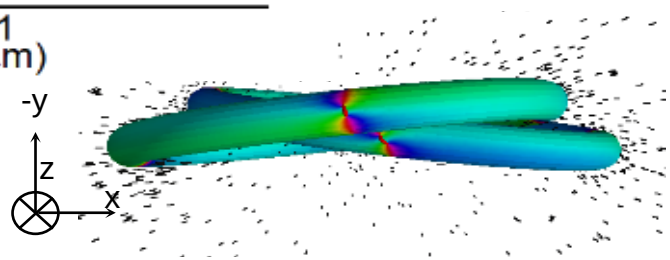
Nonlinear depth-resolved optical microscopy

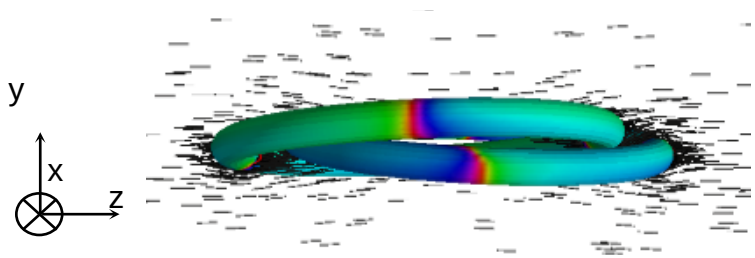


Rotational & translational elastic coupling:

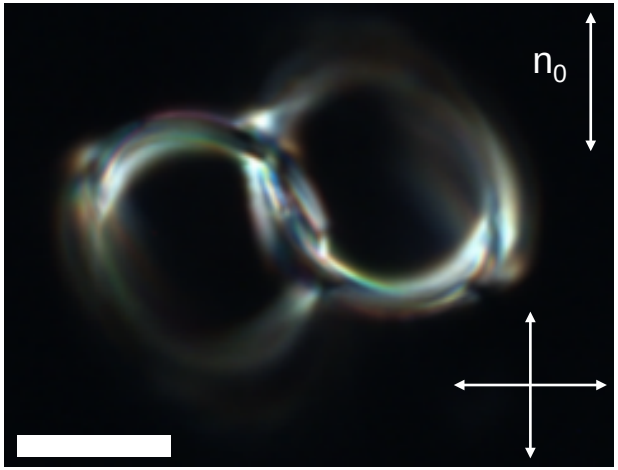


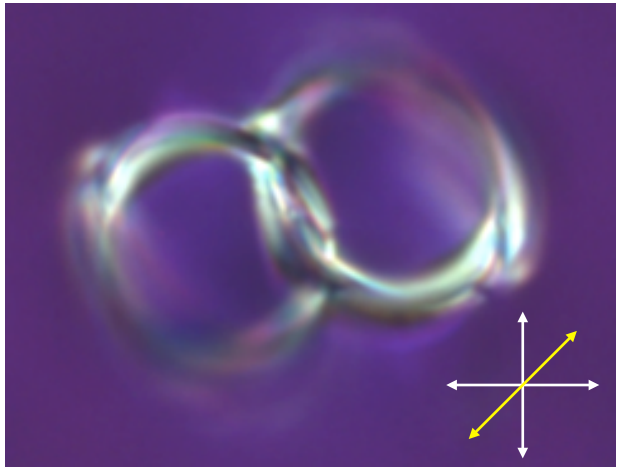


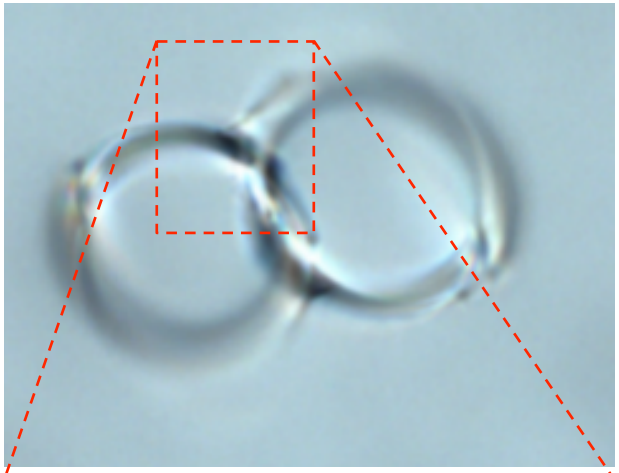


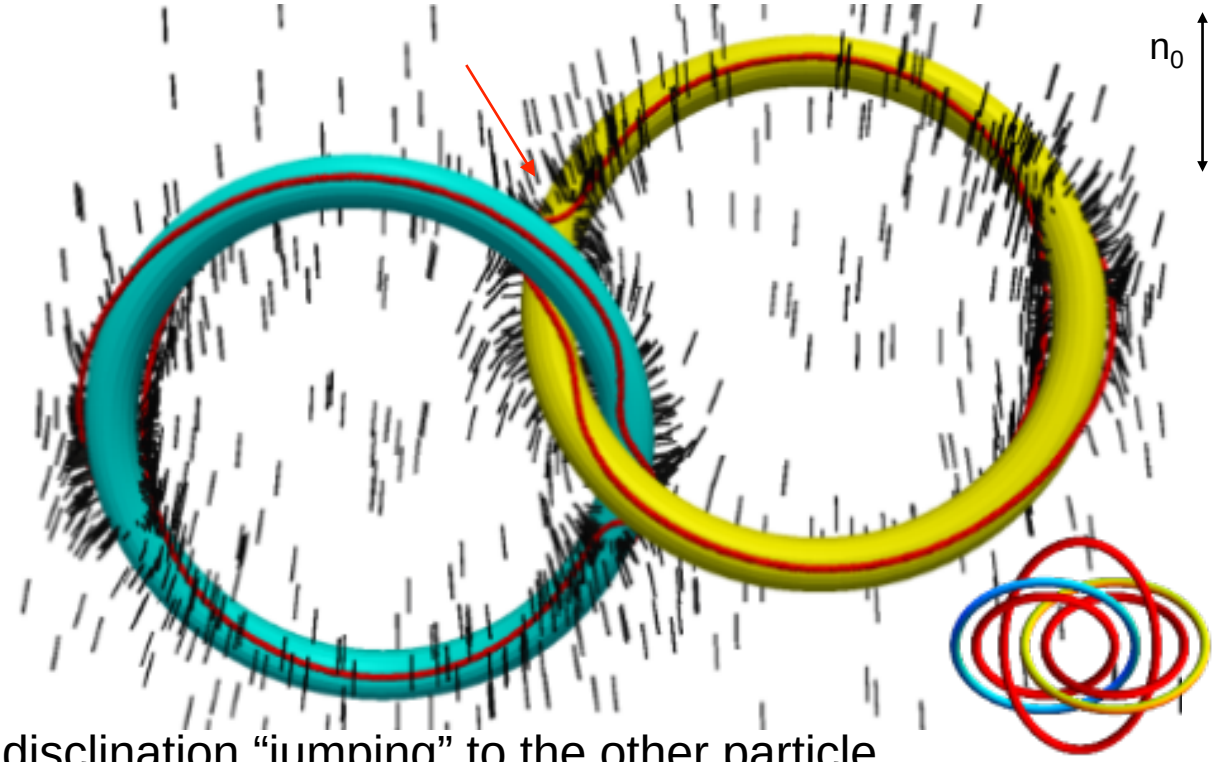


Colloidal Hopf Links with perpendicular BCs



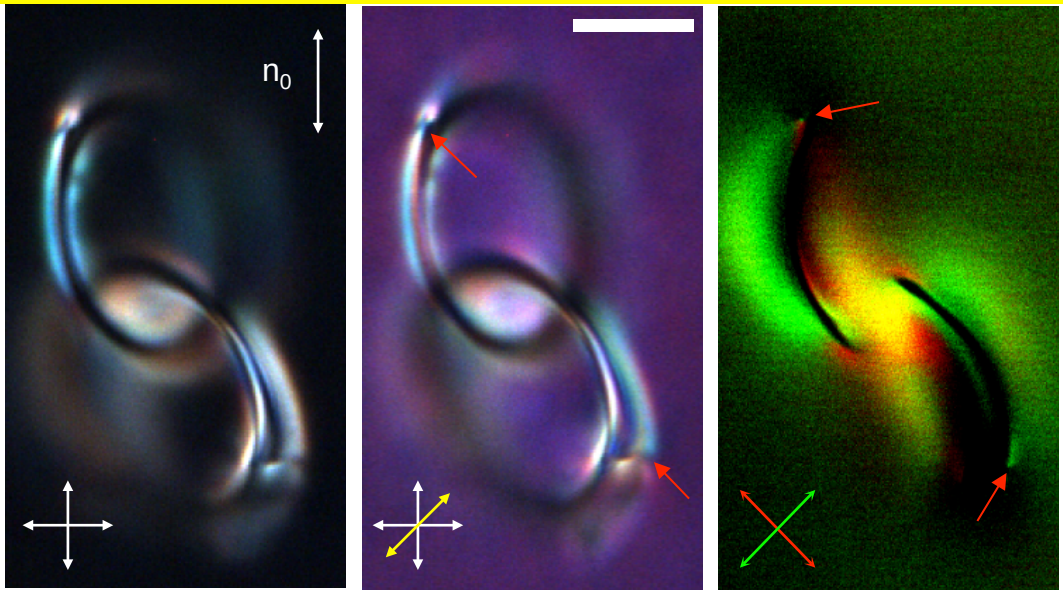


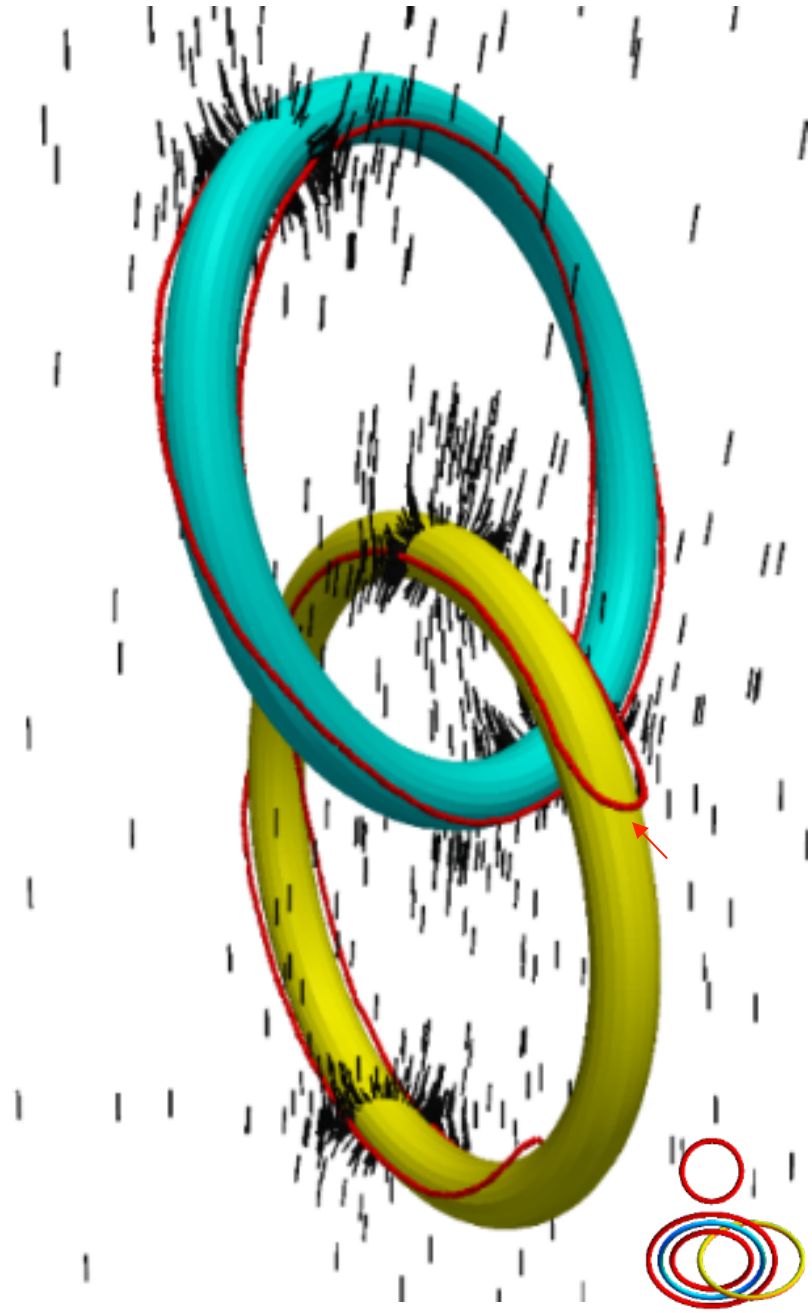




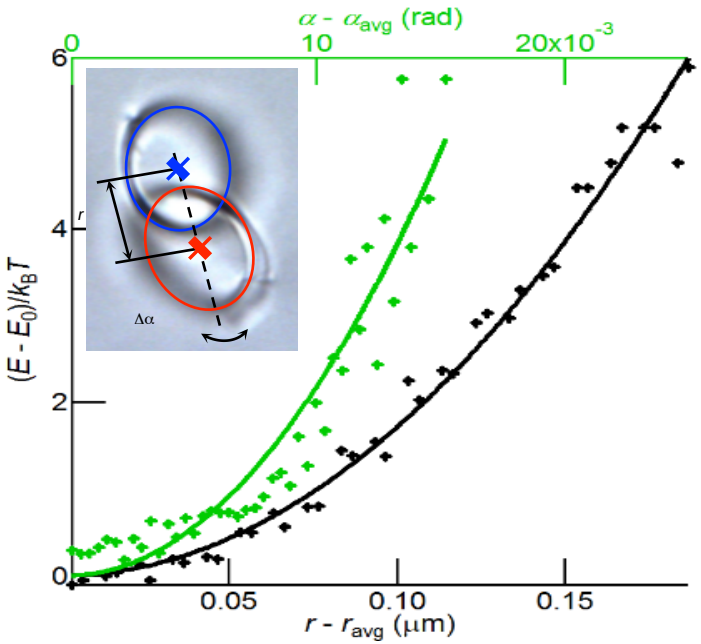
disclination “jumping” to the other particle

Metastable Hopf links with perpendicular BCs

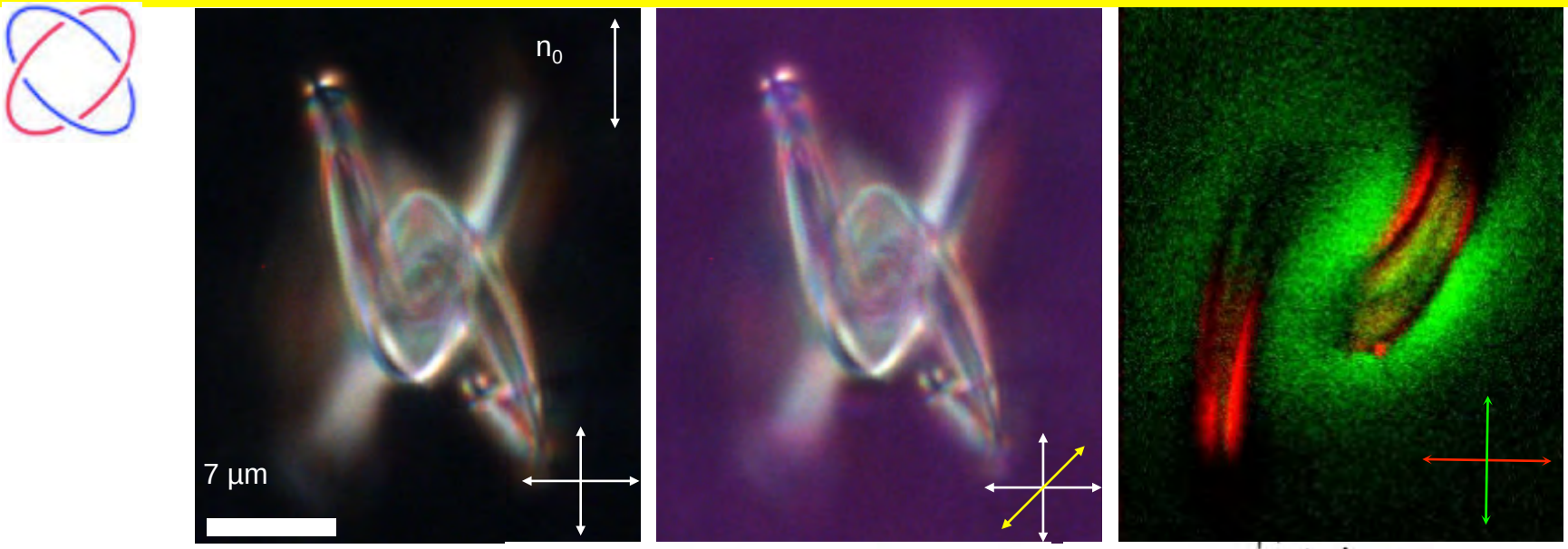




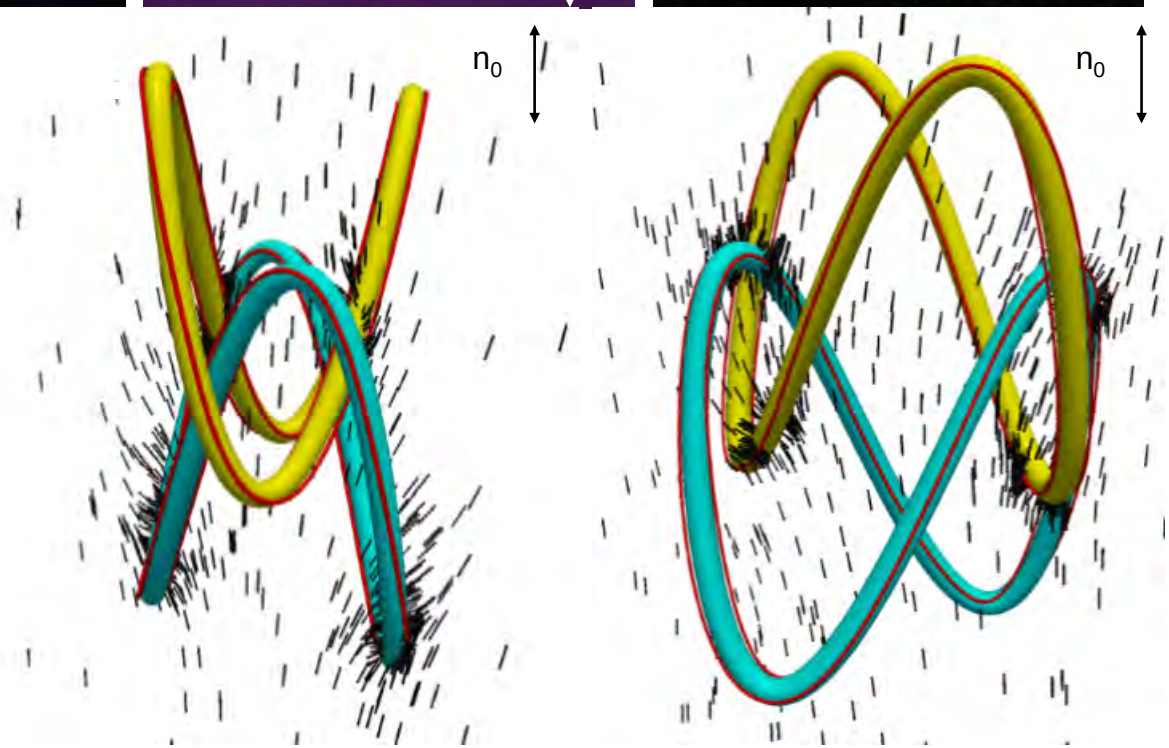
Rotational & translational elastic coupling:



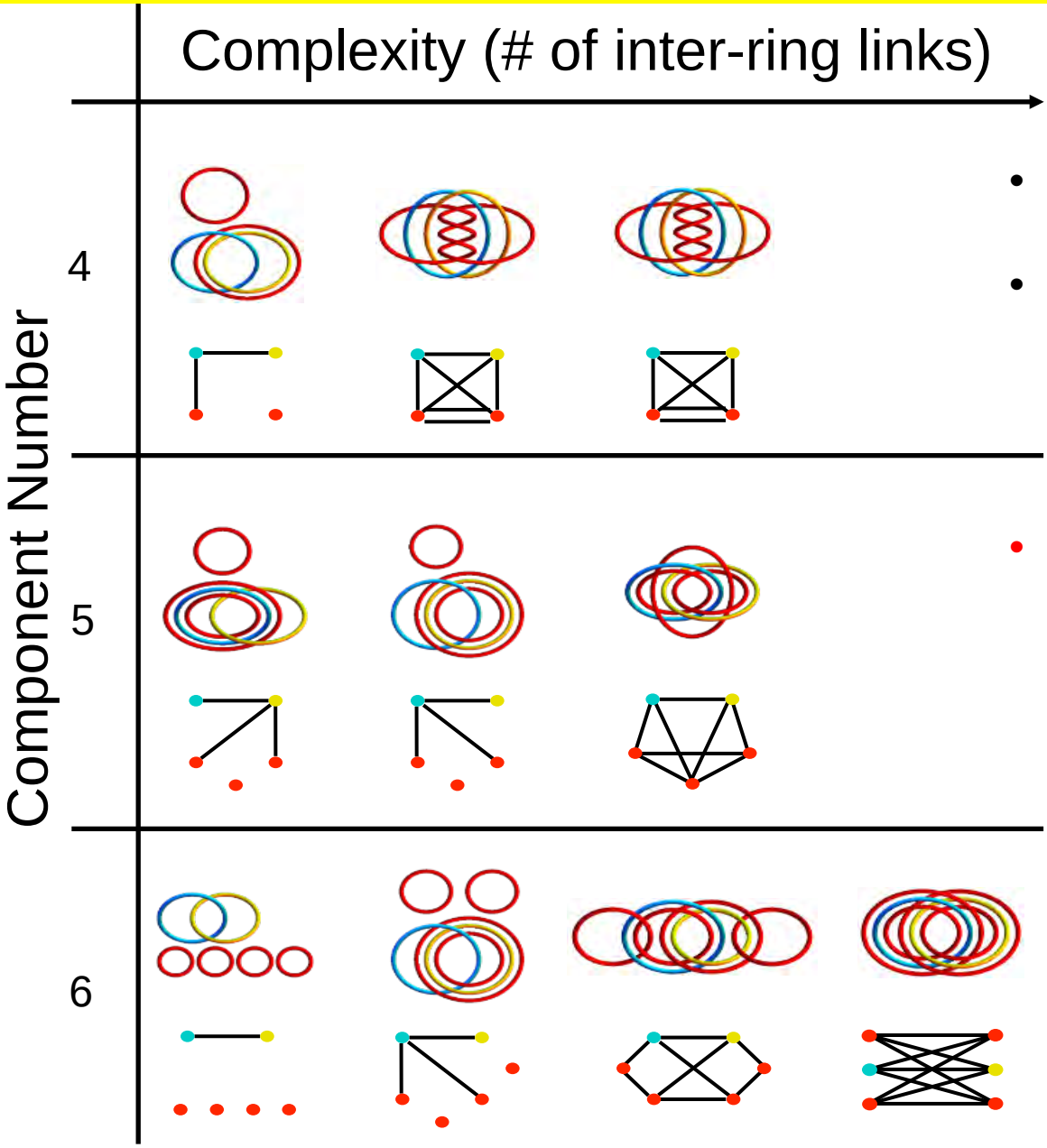
Colloidal Solomon links with homeotropic BCs



Half-integer line defects follow the particle's tubes



Summary: graph presentations of links



Homeotropic BCs:

- colloidal Hopf links accompanied by 2 or more defect rings
- defect loops have linking number of one or two

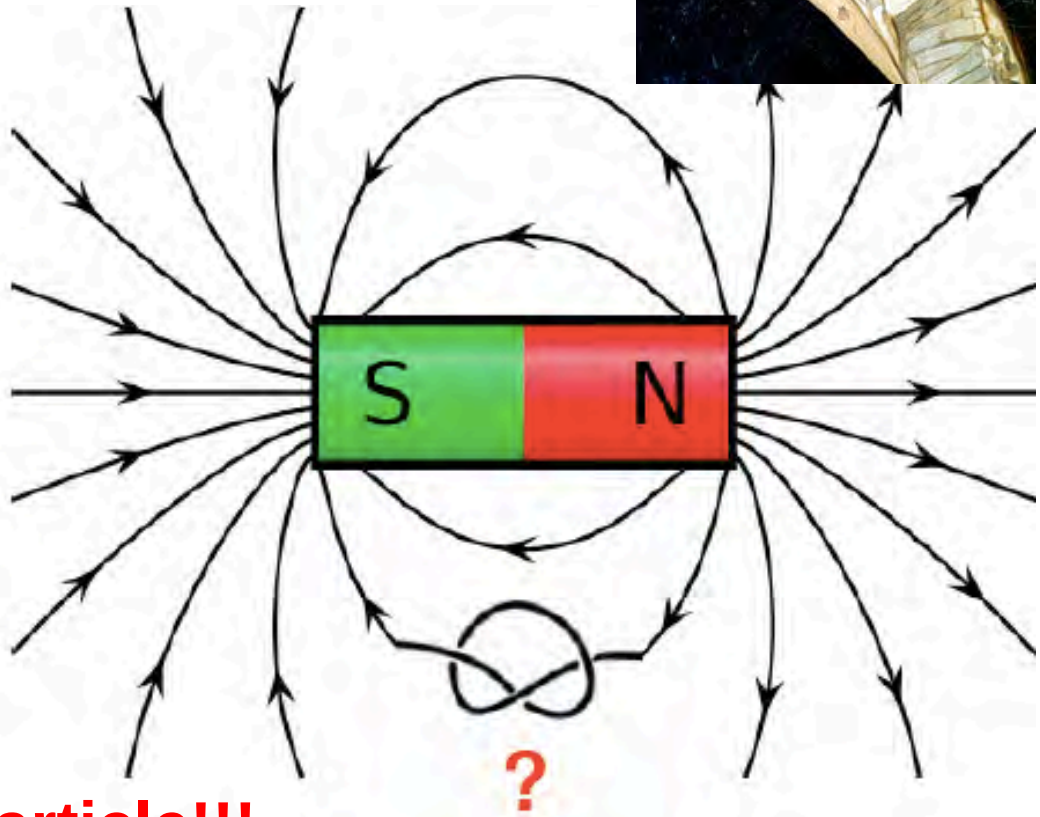
- Nonpolar nature provides many ways of satisfying topological constraints of topological interaction between fields, defects & surfaces

A. Martinez, L. Hermosillo, M. Tasinkevych, and I. I. Smalyukh. PROC. NATL. ACAD. SCI. U.S.A. 112, 4546-4551 (2015)

2D “baby” skyrmions & 3D knotted solitons (hopfions), torons, & umbilics

Links & knots in fields – particles ???

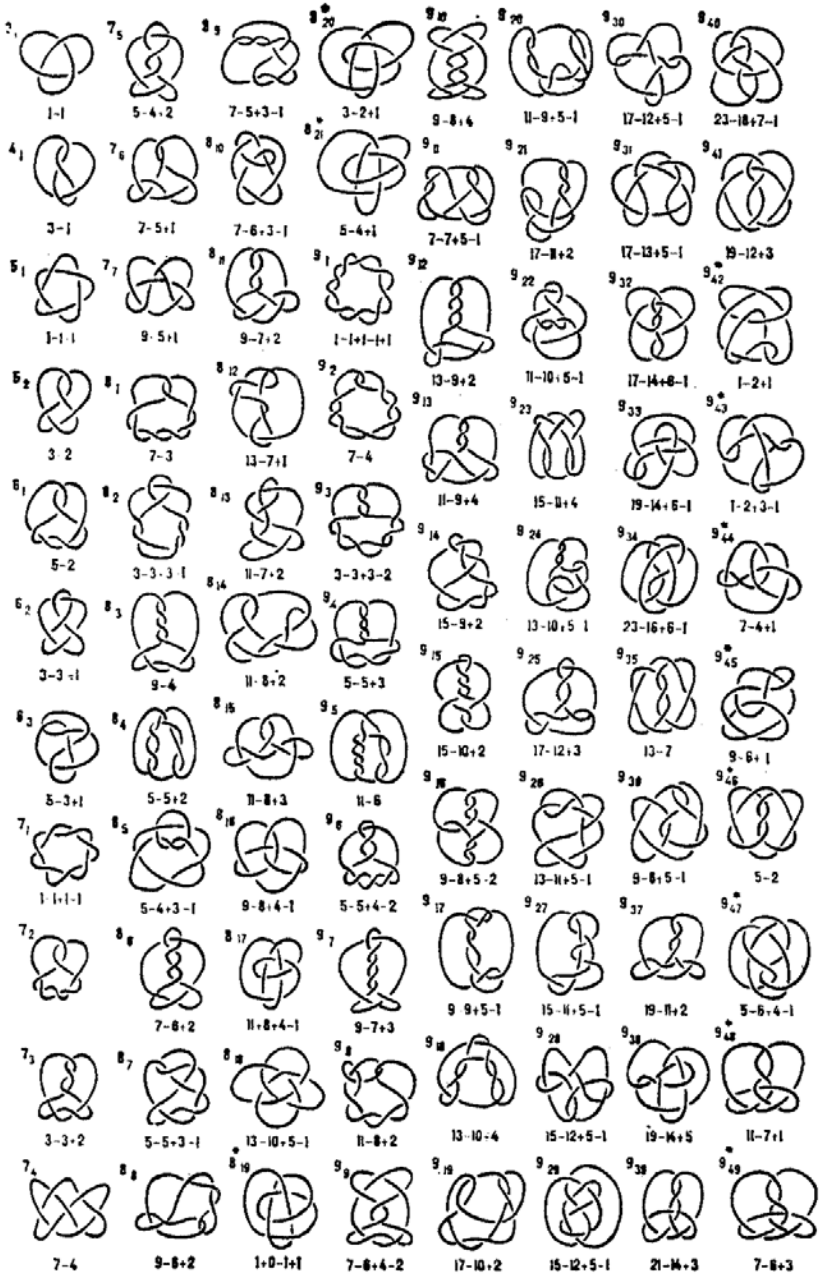
Entire field structure has to conform to the knot!!!

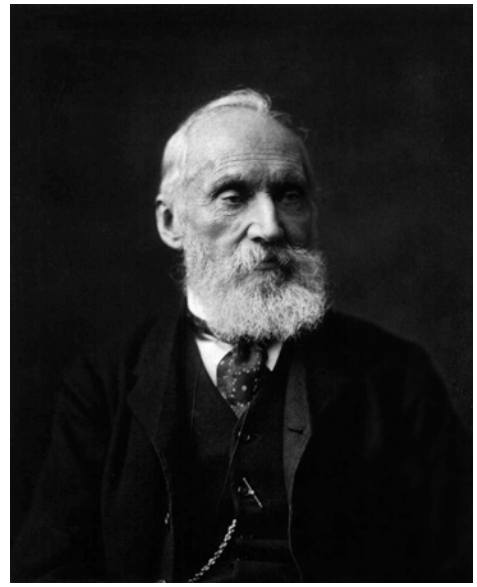


Gaus: Can behave like a particle!!!

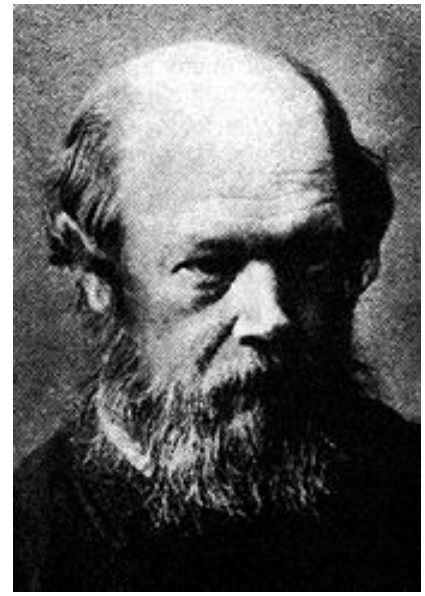
- If line crossing is prevented, knots are conserved.

Knot theory motivated by early models of atom



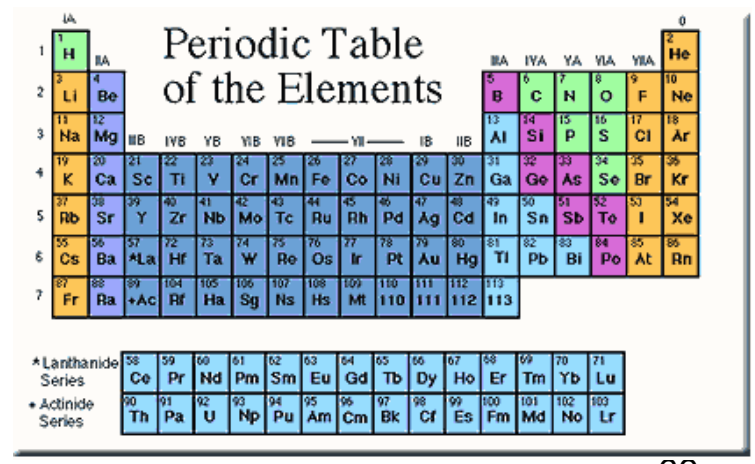


William Thomson,
Lord Kelvin



Peter Tait

$\neq$



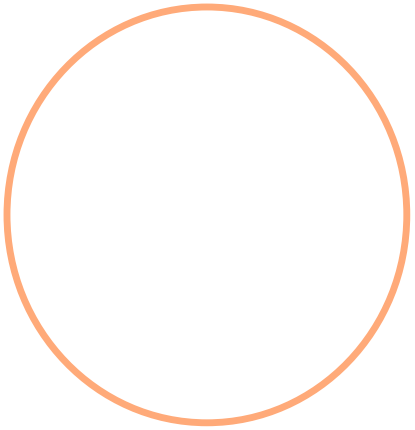
Mathematical Theory of Knots & Physics

Edward Witten

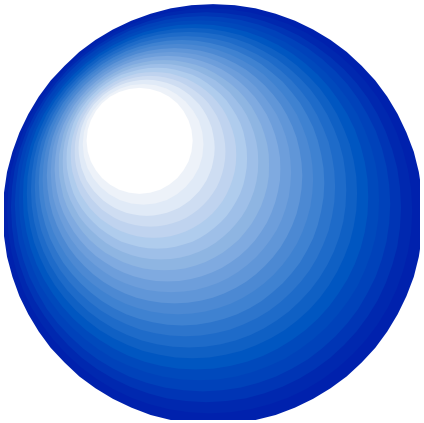
- Applications of knot theory in physics;
- Insights into the knot theory from quantum physics & field theories;
- 1990 Fields medal (the only physicist to receive);
- Knotted fields: from elementary particles to cosmology

High dimensional spheres: S^3 in 4D

S^1 in 2 Dimensions



S^2 in 3 Dimensions



S^3 in 4 Dimensions

S^2 - all points in 3D space at distance 1 from the origin.

$$\sqrt{x^2 + y^2 + z^2} = 1$$

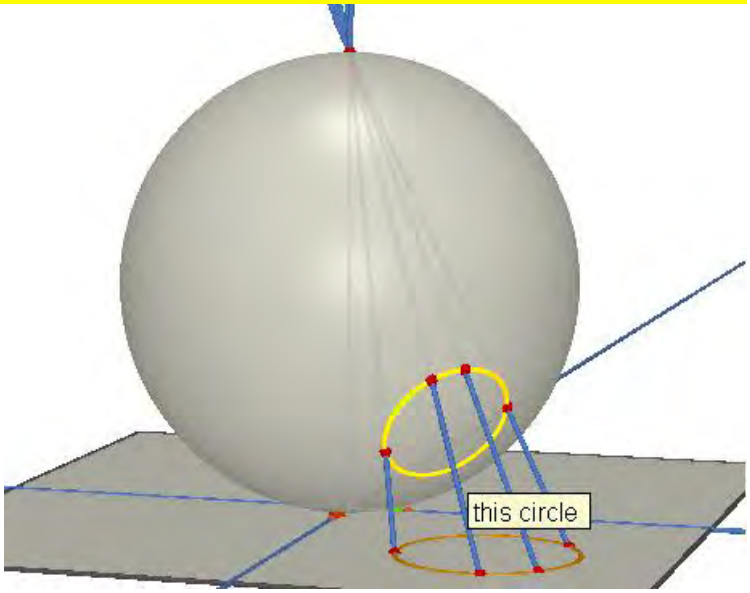
S^3 – all points (x,y,z,w) in 4D space at distance 1 from the origin.

$$\sqrt{x^2 + y^2 + z^2 + w^2} = 1$$

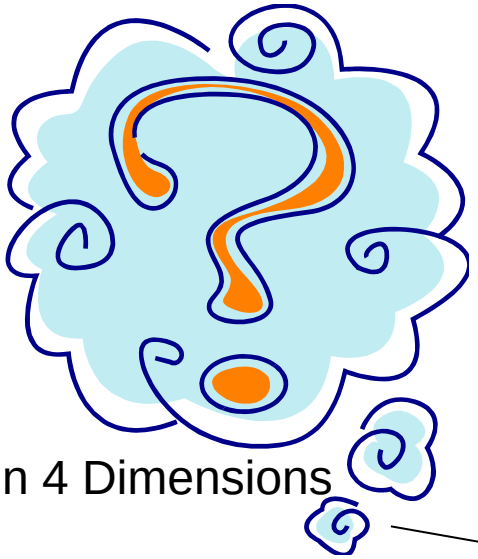
High-dimensional spheres?

Stereographic projection

- Stereographic Projection of S^2 in 3D onto a 2D plane

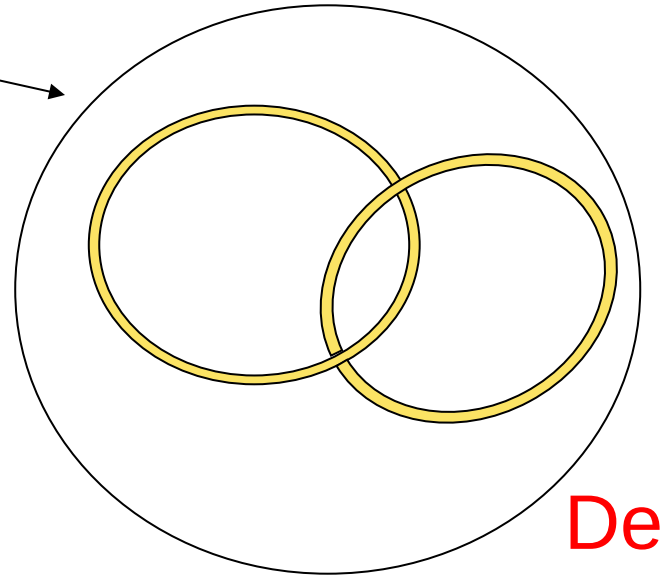


- Stereographic Projection of S^3 in 4D into the 3D space



S^3 in 4 Dimensions

Stereographic projection in 3D

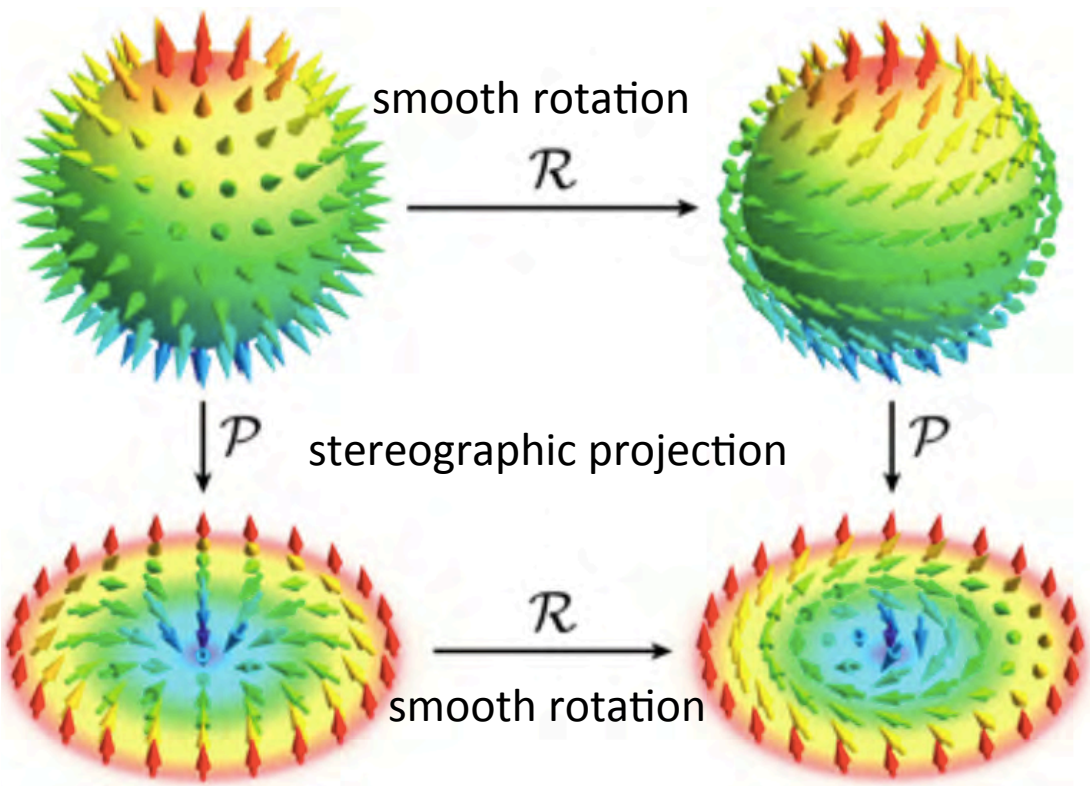


Circles are linked!

Demo by Paul

2D & 3D solitons: baby Skyrmions vs Hopfions

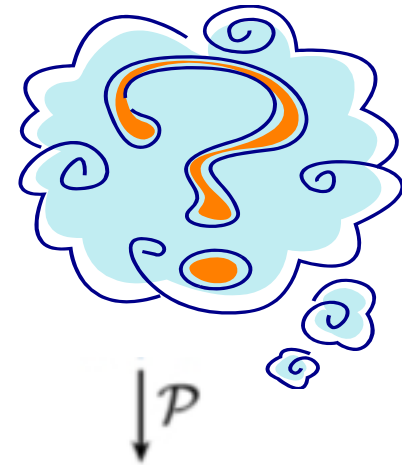
S^2 in 3 Dimensions



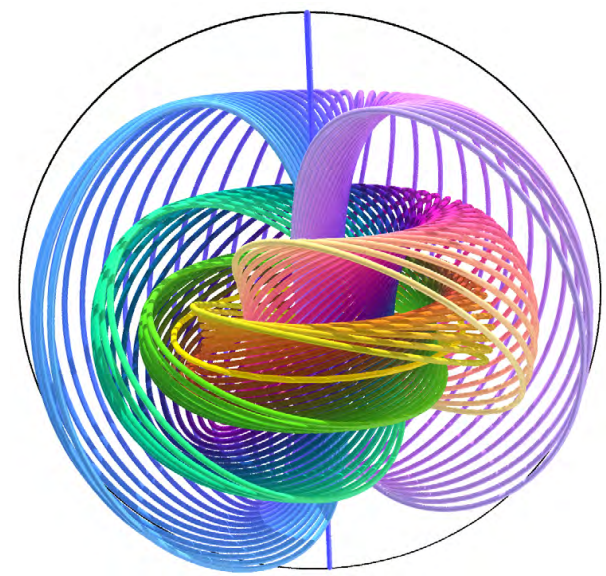
2D Soliton - ***baby*** Skyrmions in chiral magnets & in chiral liquid crystals

Topology-protected solitons!

S^3 in 4 Dimensions

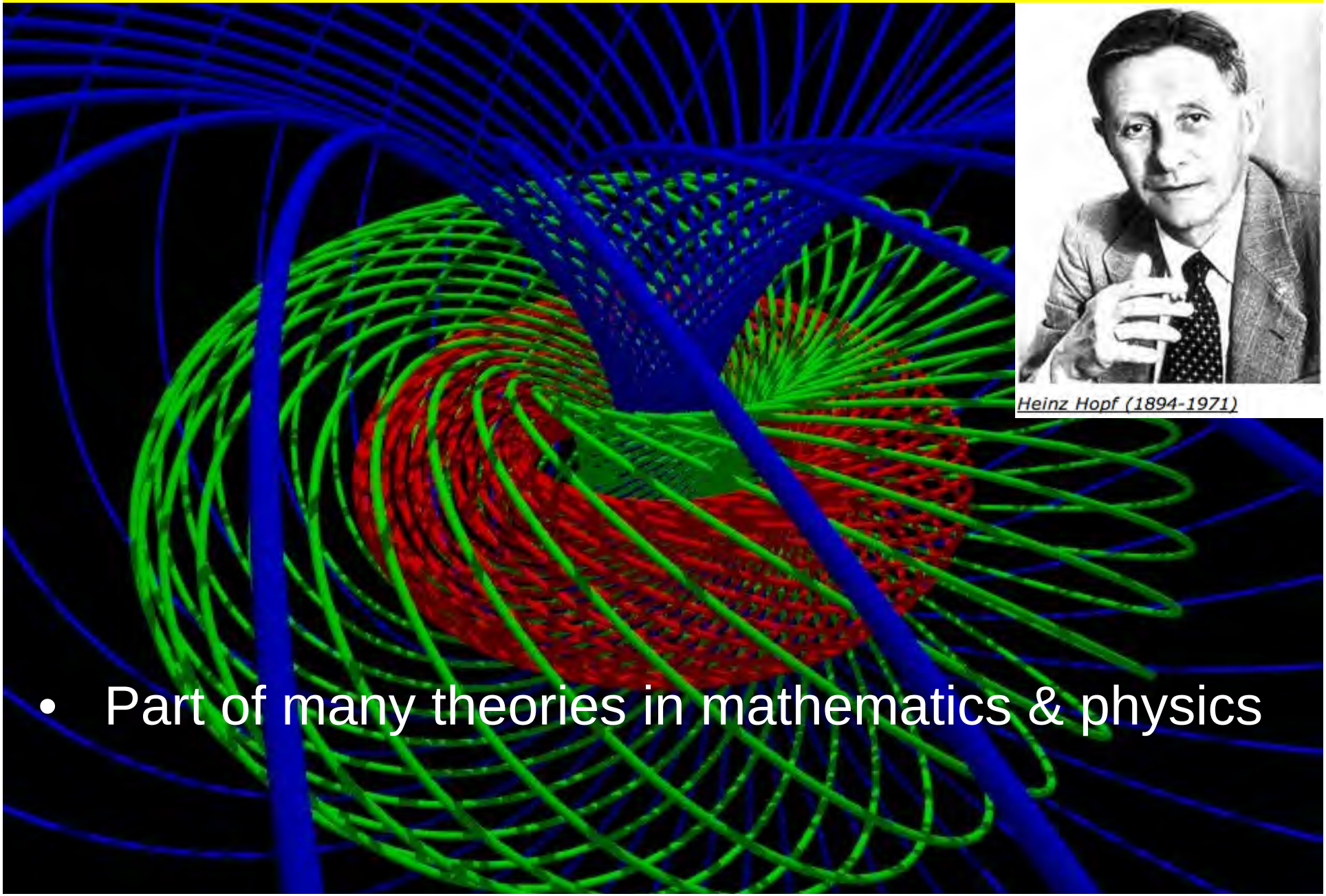


$\mathcal{P}$



3D Soliton – Hopfion in chiral LCs (3D Skyrmions)

Mathematical Hopf fibration

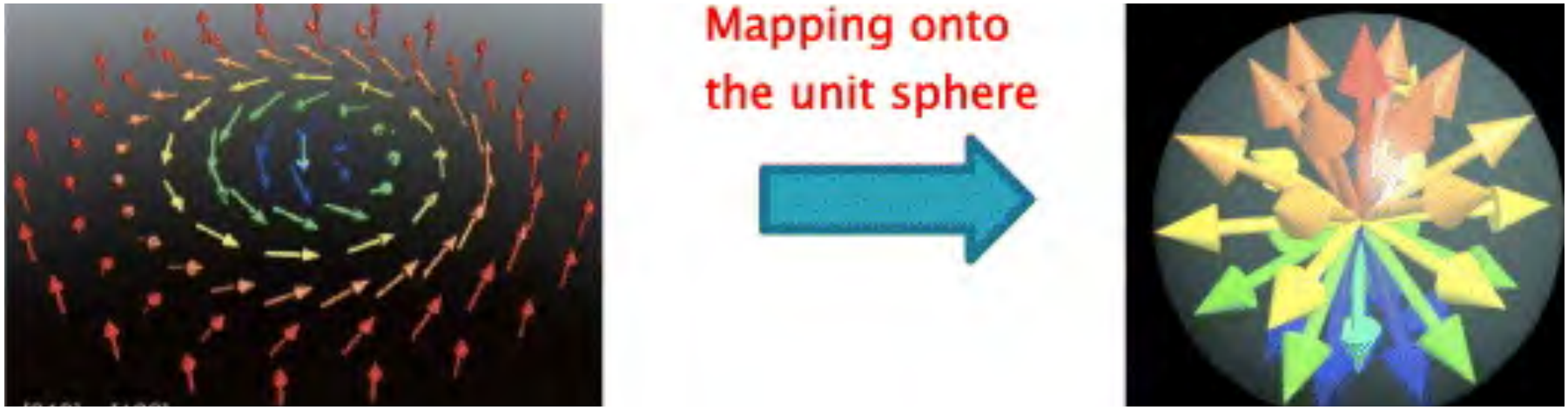


Heinz Hopf (1894-1971)

- Part of many theories in mathematics & physics

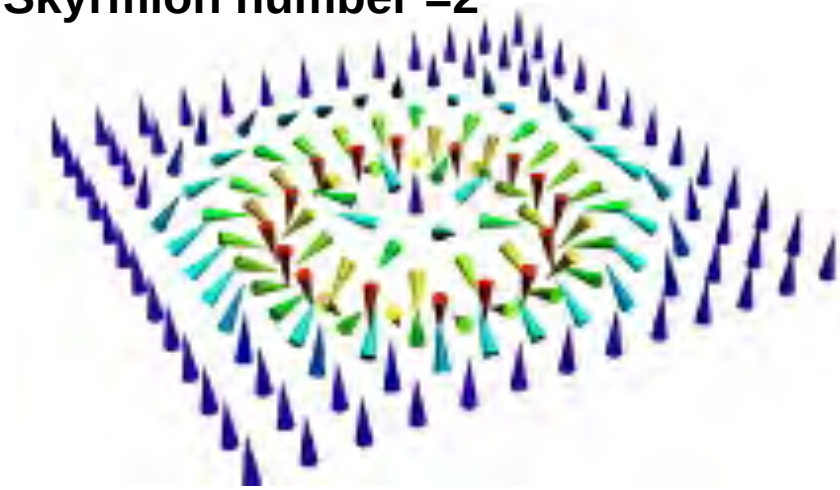
Nonsingular configurations: Skyrmions, “Baby” Skyrmions…

Skyrmion



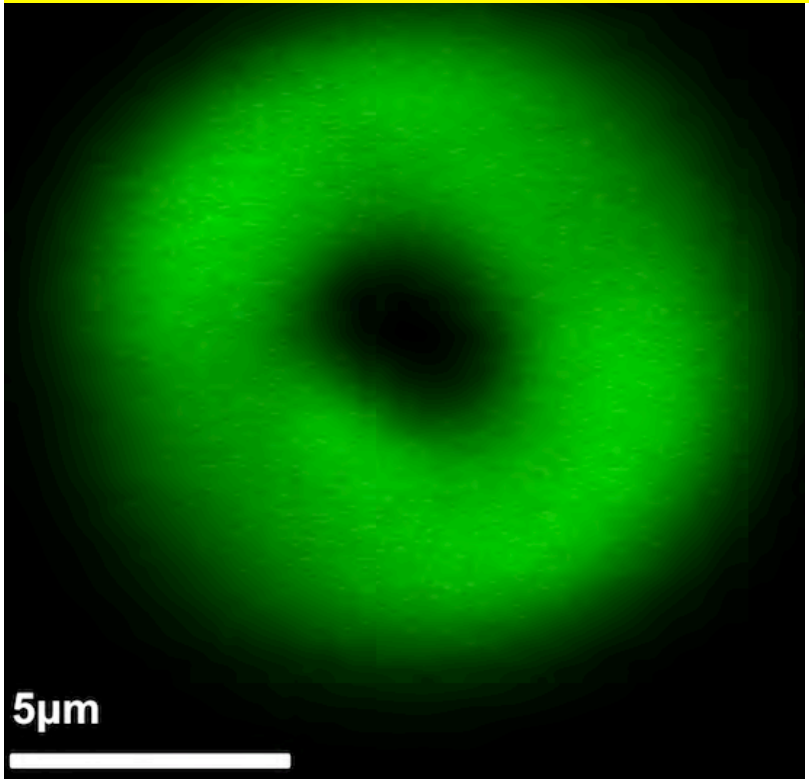
Skyrmion number - how many times the mapping wraps the unit sphere.

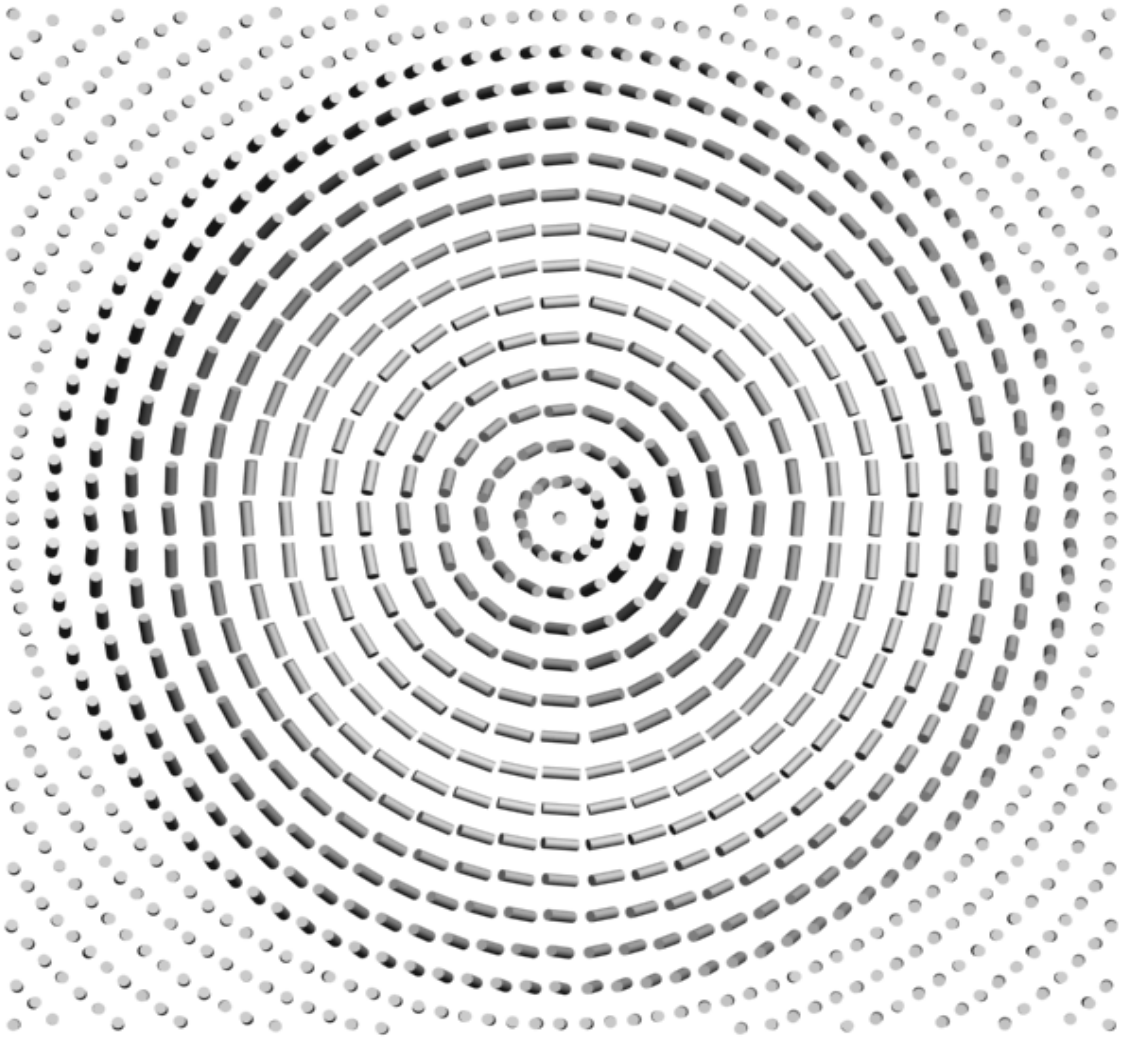
Skyrmion number =2



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

“Baby Skyrmion” in a confined chiral nematic



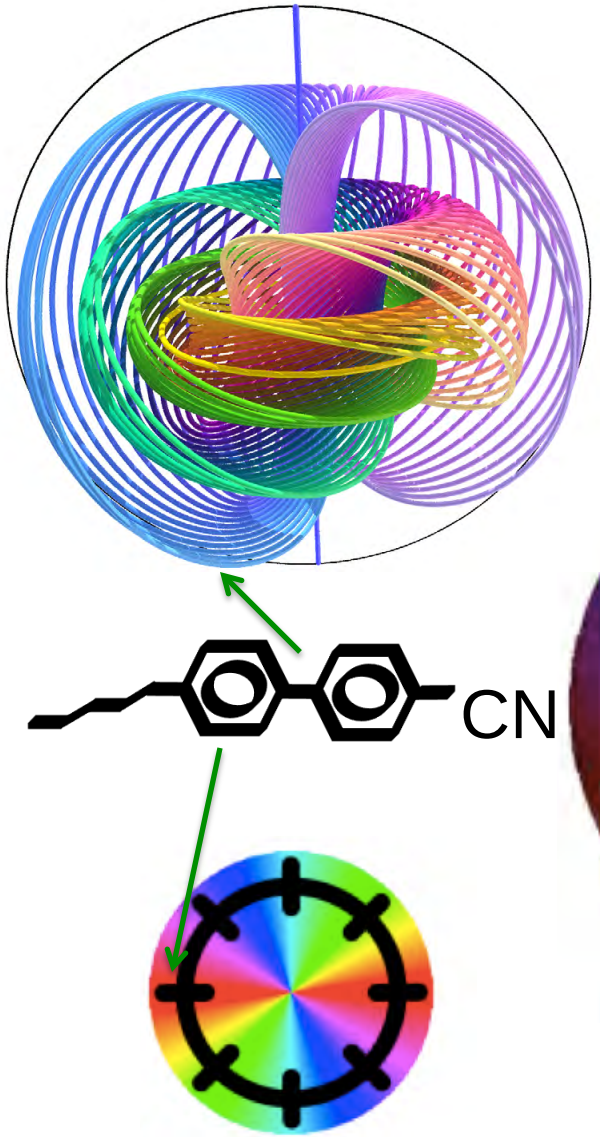


$$\pi_2(\mathbb{R}P^2) = \mathbb{Z}$$

- Double-twist cylinder
- Translationally invariant in direction orthogonal to the screen

P. J. Ackerman, R. P. Trivedi, B. Senyuk, J. van de Lagemaat, and I. I. Smalyukh. Phys Rev E **90**, 012505 (2014)

Hopf Fibration in Molecular Orientation Field

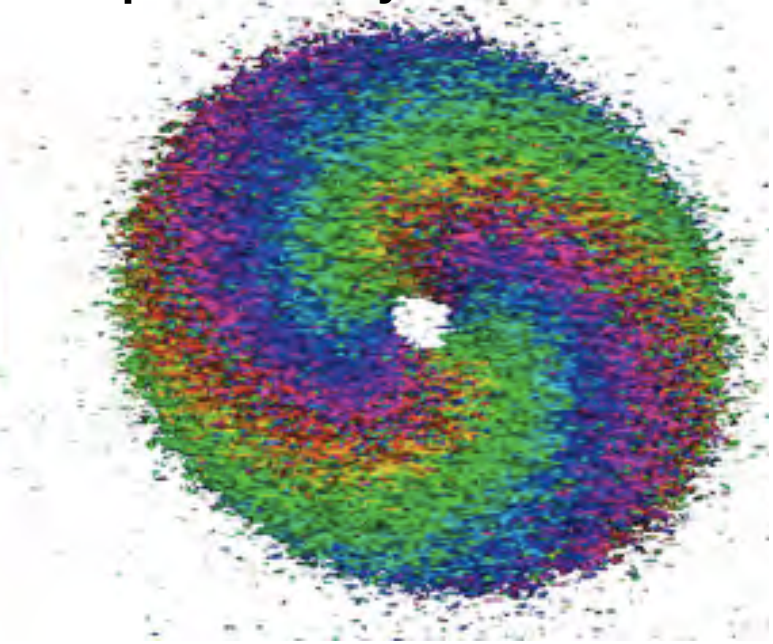


Color-coded molecular orientation field

Pontryagin-Thom construction:

computer-simulated

experimentally reconstructed



Smalyukh et al. Nature Materials 9, 139-145 (2010).
Chen, Ackerman, Alexander, Kamien, Smalyukh Phys. Rev. Lett. **110**, 237801 (2013)

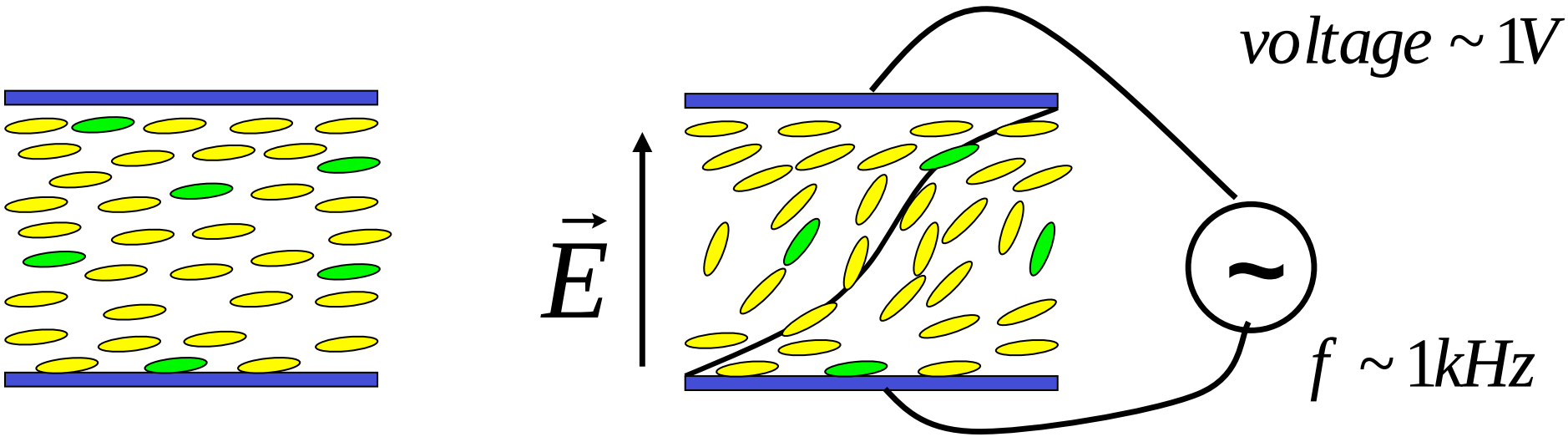
Electrical realignment

→**Facile response;**

→**Field-induced realignment;**

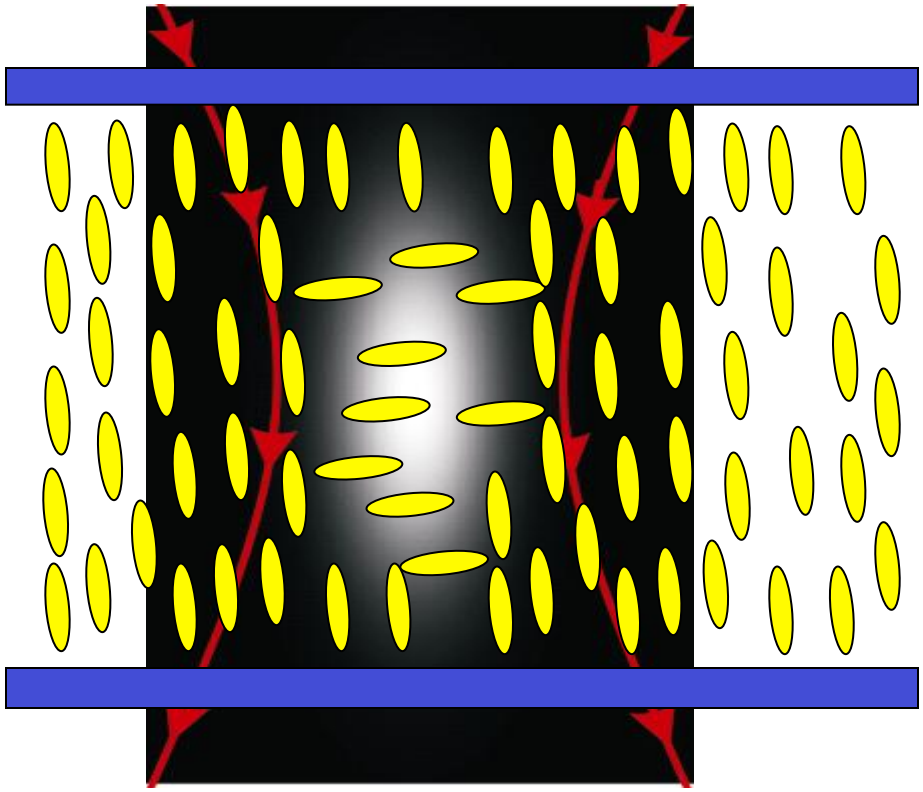
→**Minimize the electric-field term of free energy density:**

$$F_e \propto -(\vec{E} \cdot \hat{n})^2$$



Optical realignment

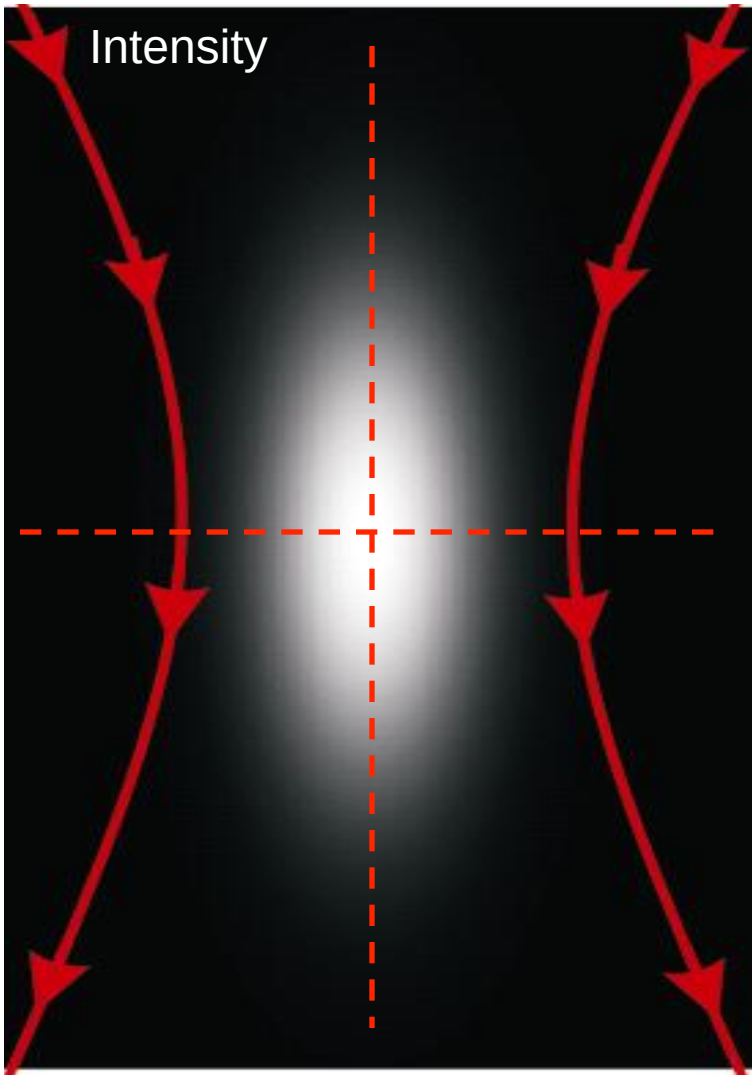
Optical realignment



→Realignment by field of laser beam;

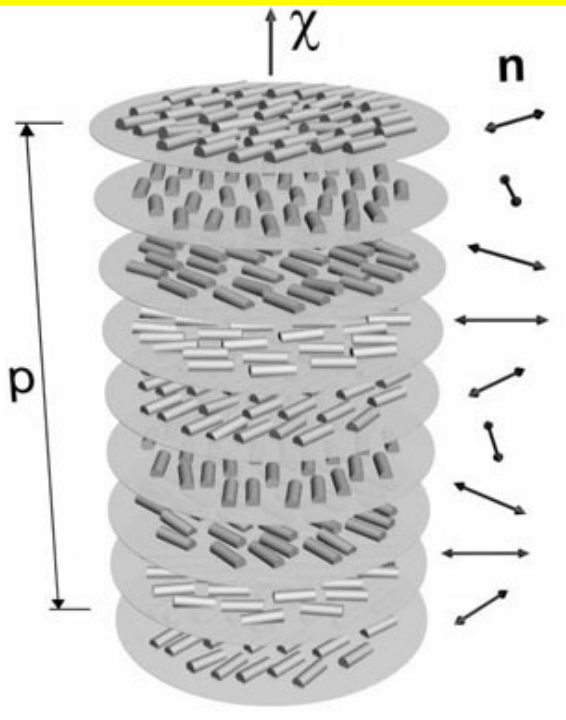
$$F_e \propto -(\vec{E} \cdot \hat{n})^2$$

Focused Gaussian beam



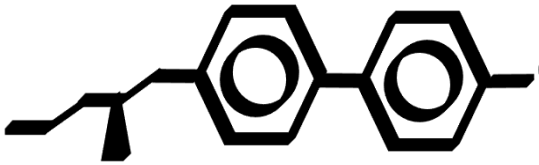
$$Intensity \sim E^2$$

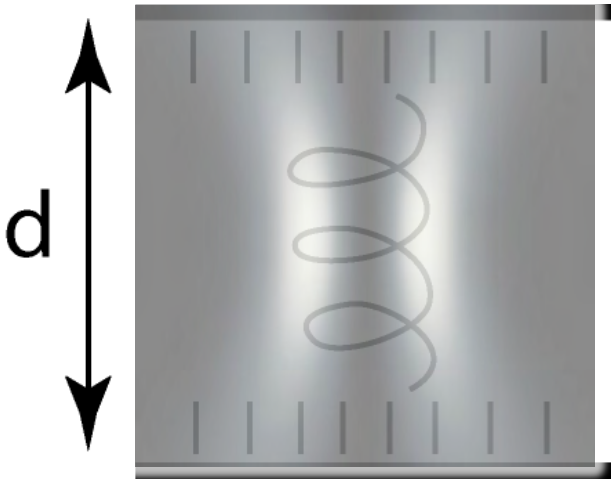
Chiral nematic & vertical boundary conditions



5CB  CN

Chiral LC molecule :

CB15  CN



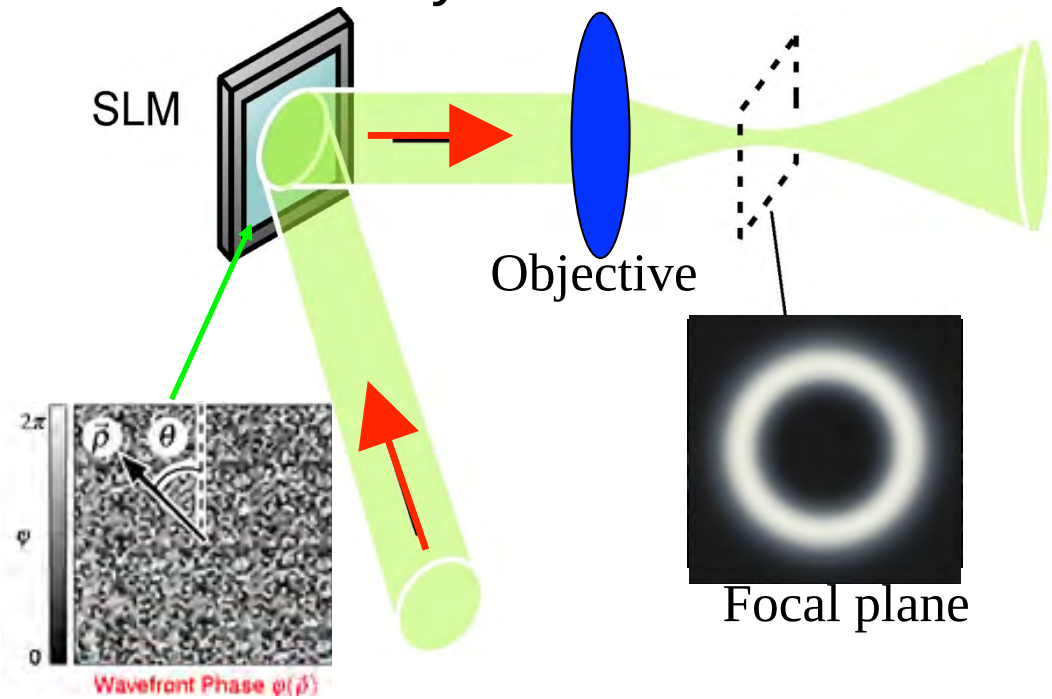
→ Vertical BCs incompatible with twist

→Result – frustration & unwinding

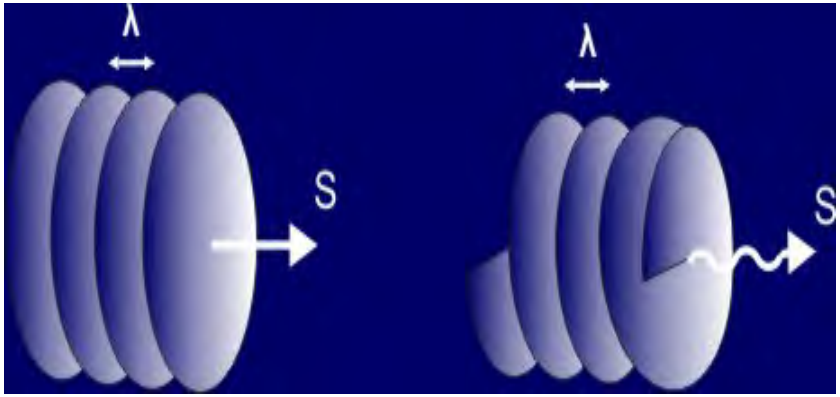
LG beam's effect?

Beam shaping using a spatial light modulator

Phase-only LC SLM

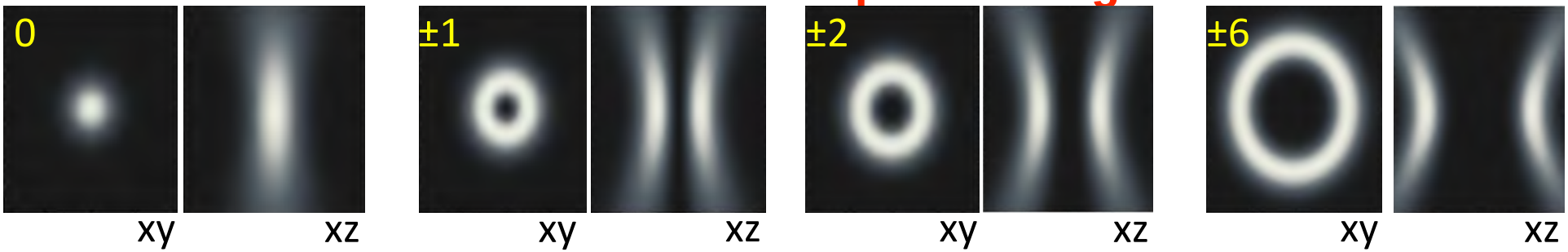


Optical vortices - phase singularities - screw dislocations



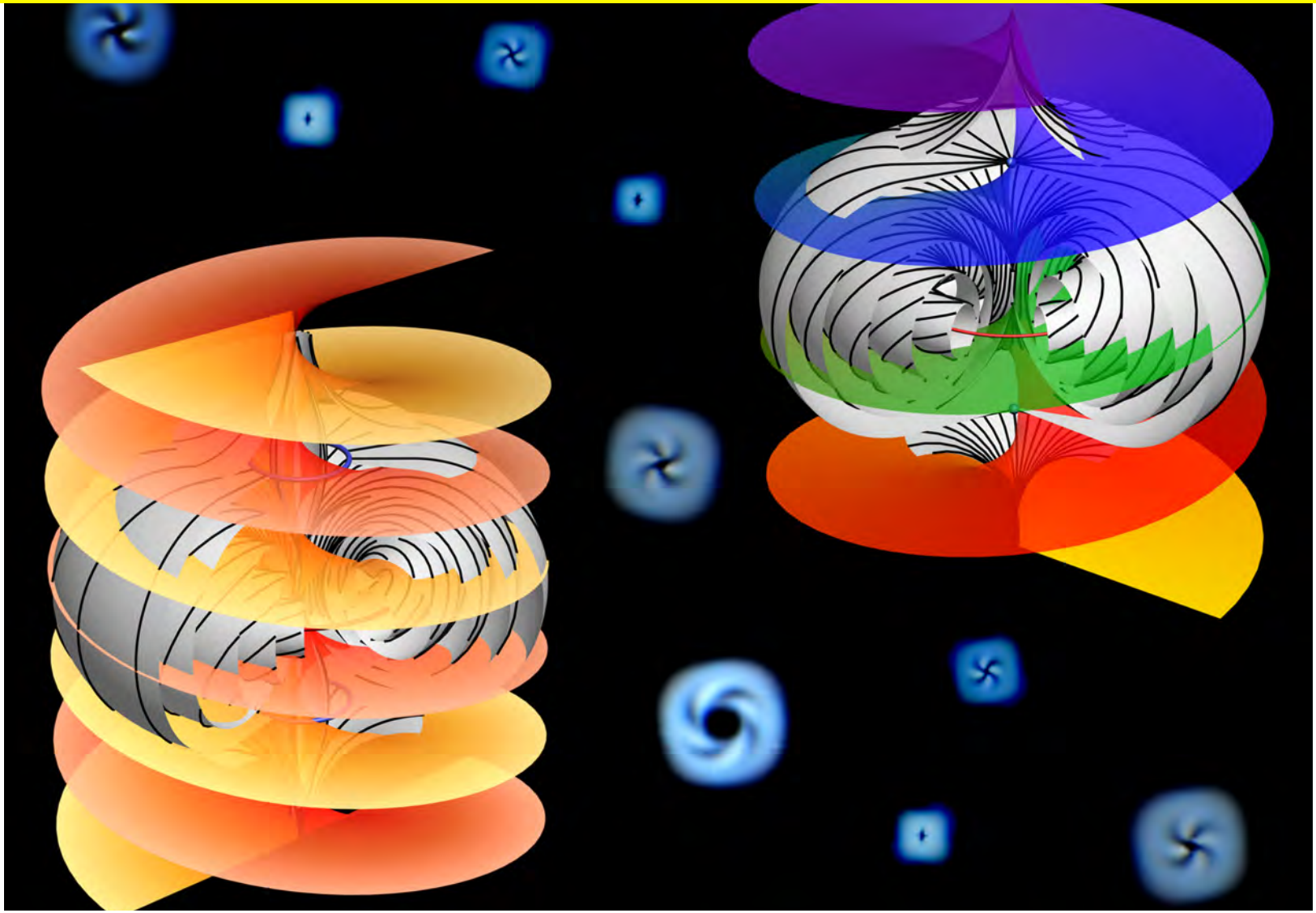
*L. Allen et al., PRA **45**, 8185 (1992)*

•Screw dislocations in the phase of light



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

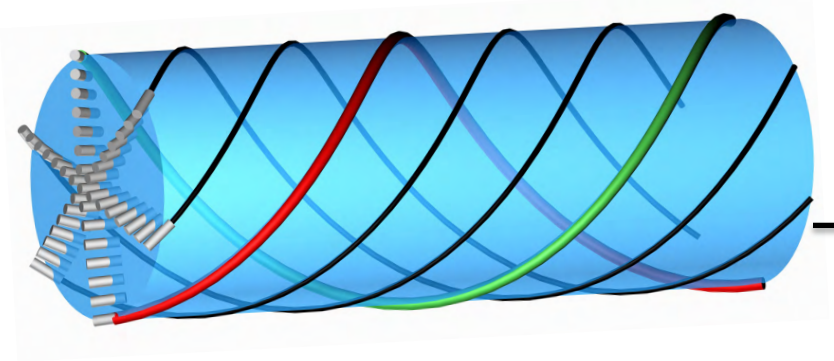
Control of topological solitons with optical vortices



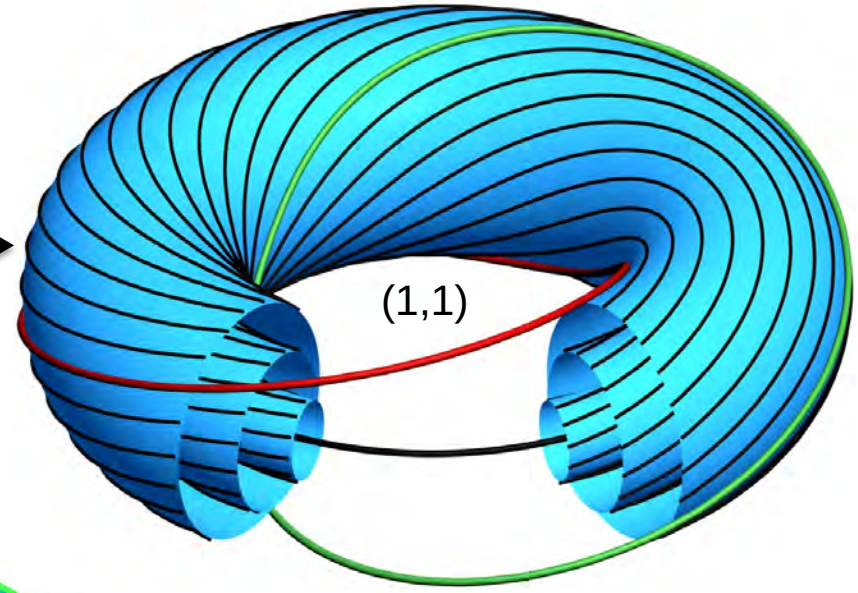
- Focus 50mW LG beams into a chiral nematic for ~10ms;
- Solitons form spontaneously – different depending on LG charge;
- Stable after turning off the laser light.

3D solitons “Hopfions”: torus knots of nematic field

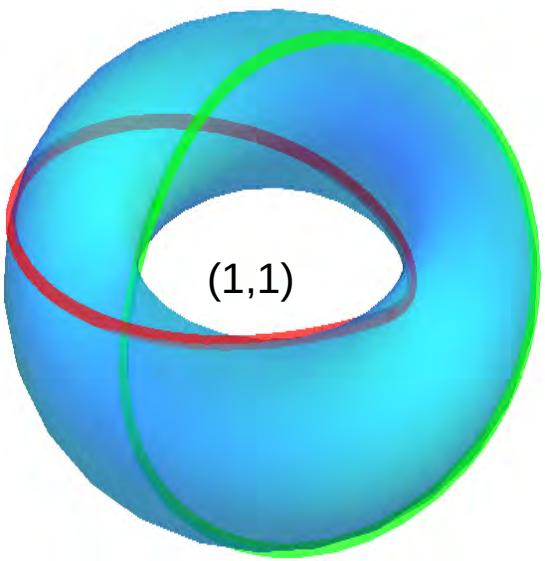
Double-twist cylinder (recall blue phases)

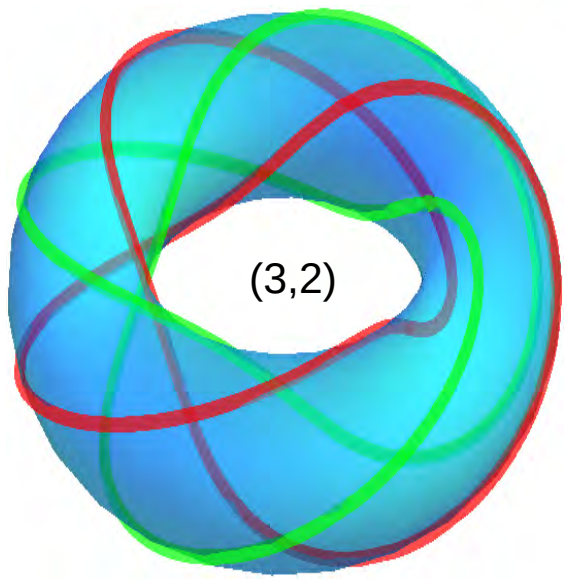


Double-twist torus



Different (P,Q)-torus knots

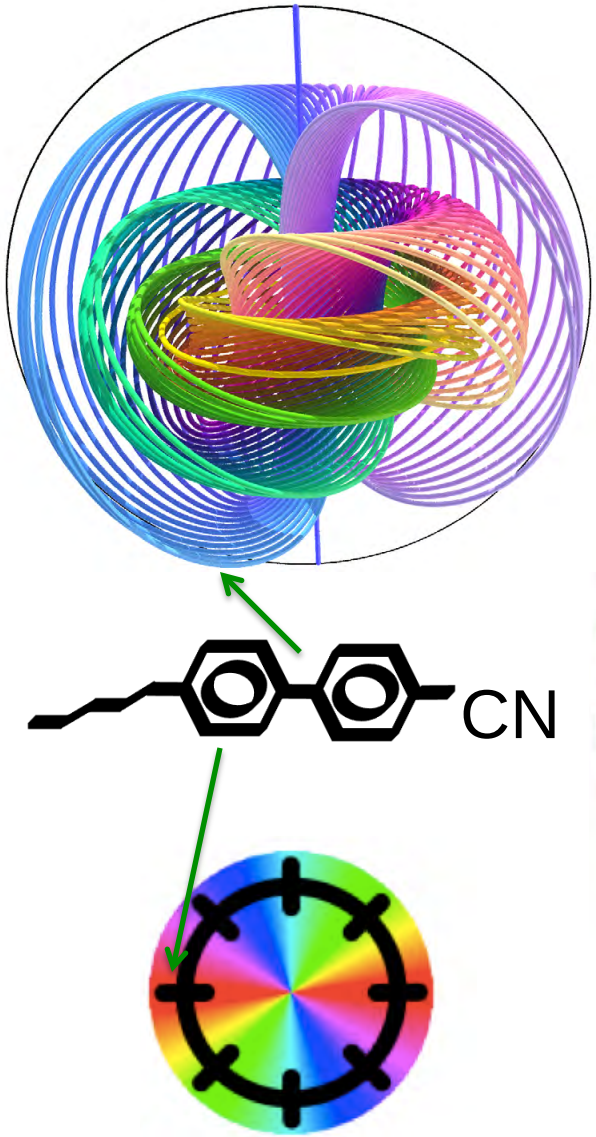




→Realignment by E-field of vortex laser beams of different charge;
→Hopf & Seifert fibrations

Hopf index: linking interpretation

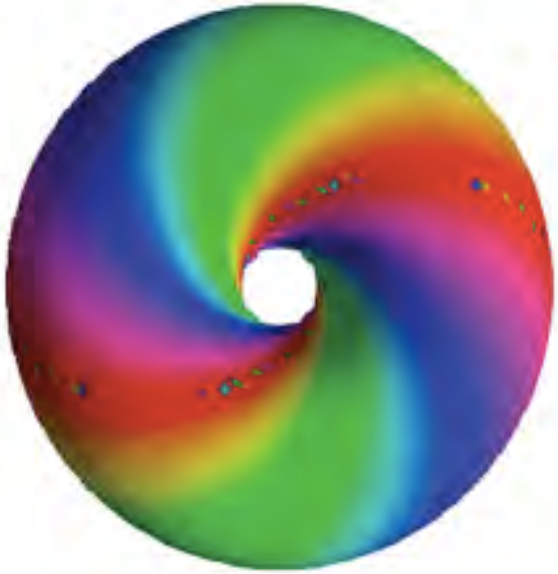
Hopf Fibration in Molecular Orientation Field



Color-coded molecular orientation field

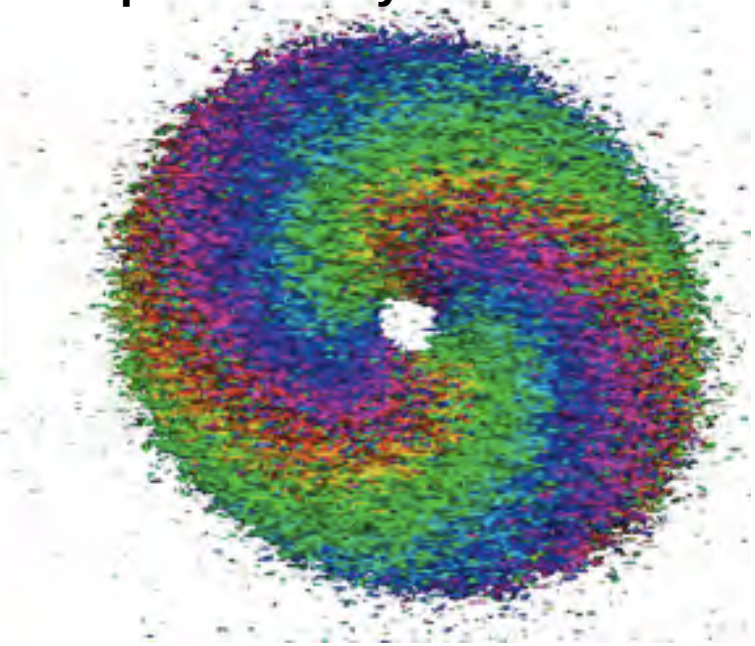
Pontryagin-Thom reconstruction:

computer-simulated



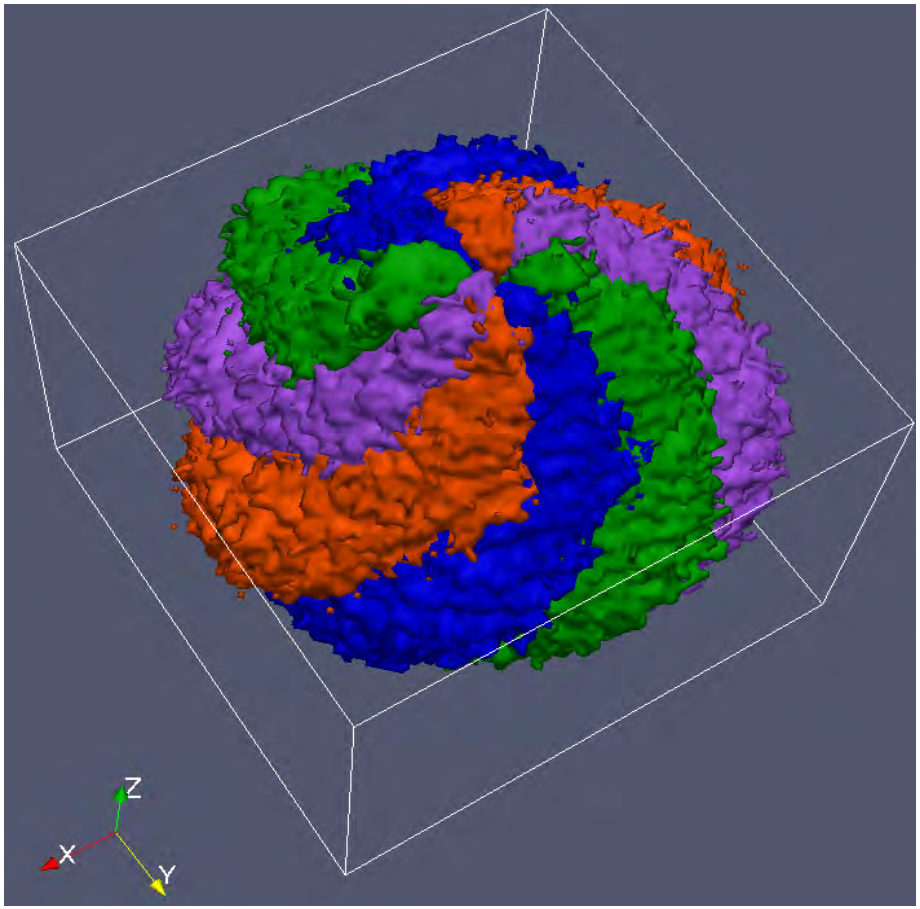
Hopf index H=1

experimentally reconstructed

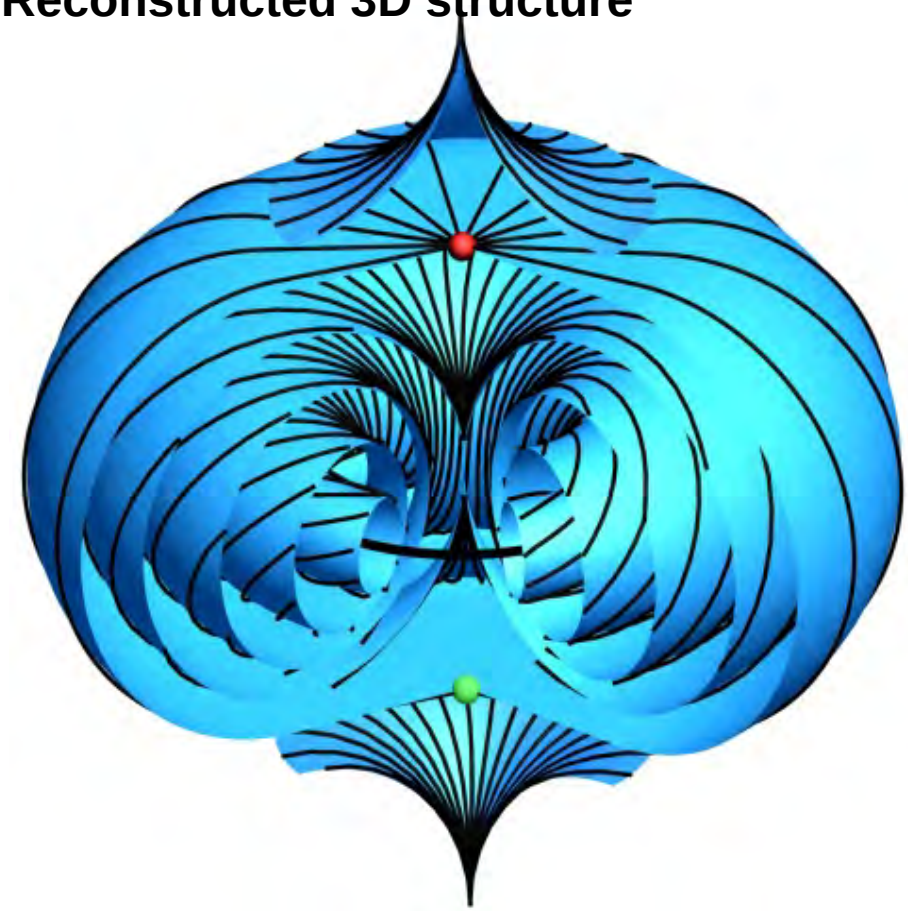


$$\pi_3(\mathbb{R}P^2) = \mathbb{Z}$$

Optically-generated Toron: Hopf + defects



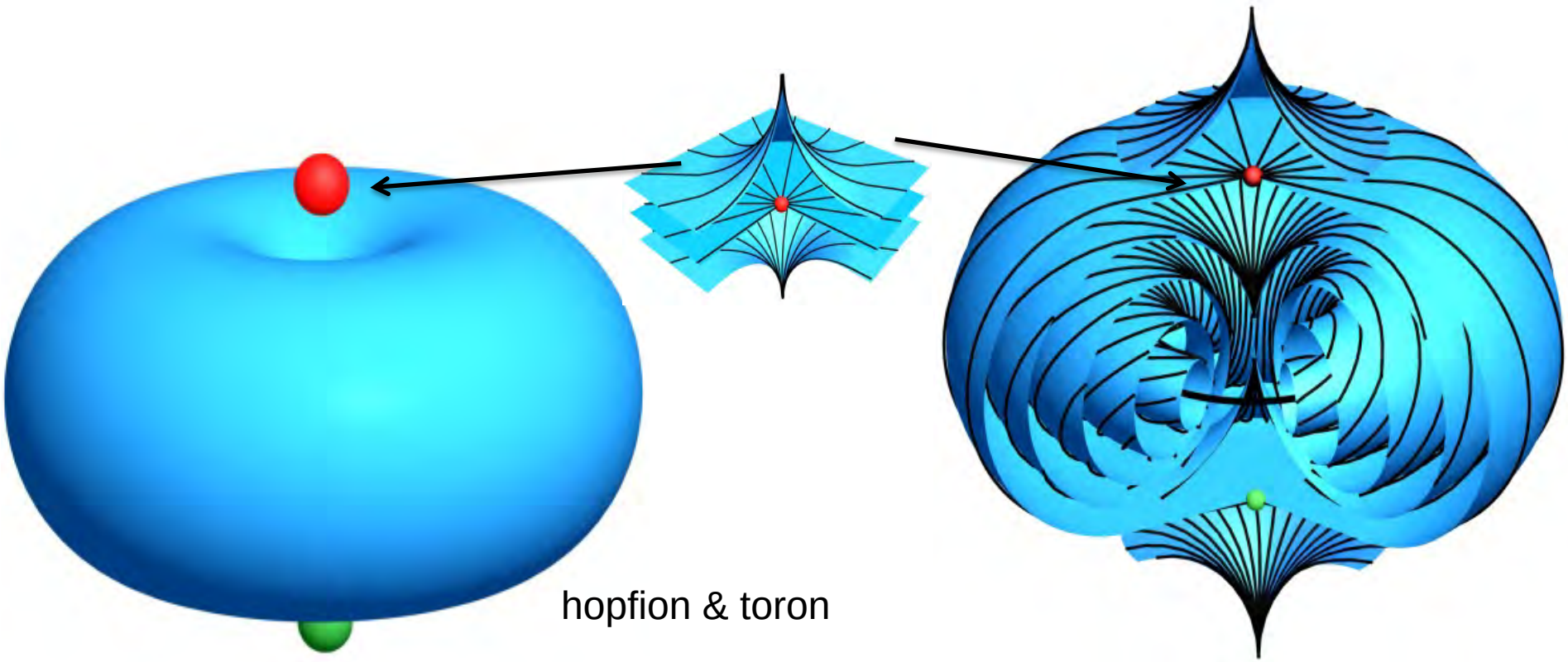
Reconstructed 3D structure



- Focus a 50mW LG beam into the chiral nematic for 10-50ms;
- Toron forms spontaneously;
- Stable after turning off the laser light.

I. Smalyukh *et al. Nature Materials* **9**, 139-145 (2010).

Optically-generated Toron: Hopf + defects



- Focus a 50mW LG beam into the chiral nematic for 10-50ms;
- Toron forms spontaneously;
- Stable after turning off the laser light.

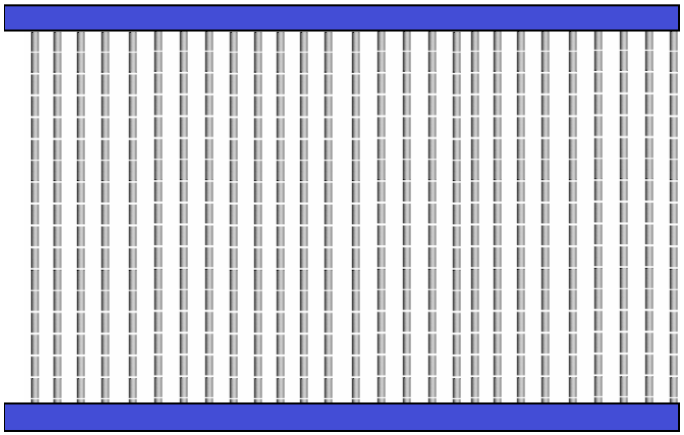
I. Smalyukh *et al. Nature Materials* **9**, 139-145 (2010).

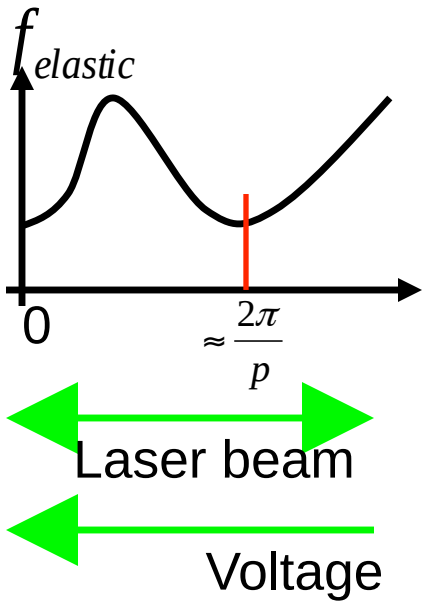
Twist vs. no twist in a “frustrated” chiral LC

Splay  Twist  Bend 

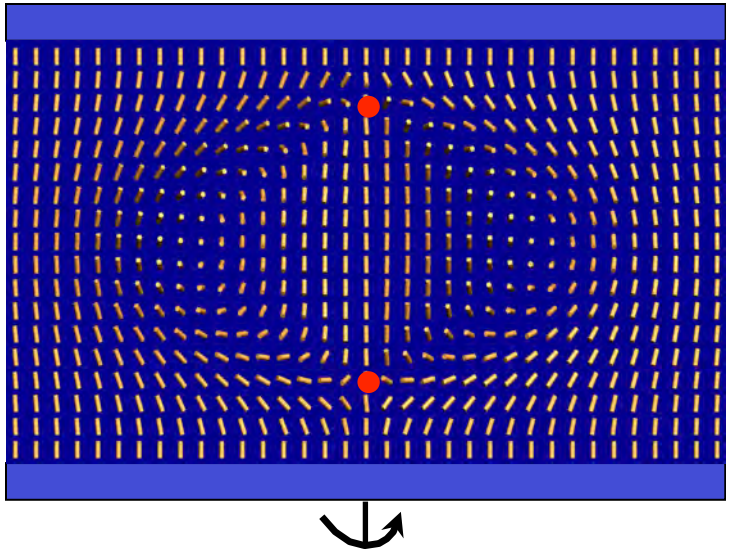
$$f_{elastic} = \frac{K_{11}}{2}(\nabla \cdot \hat{n})^2 + \frac{K_{22}}{2}[\hat{n} \cdot (\nabla \times \hat{n}) + \frac{2\pi}{p}]^2 + \frac{K_{33}}{2}[\hat{n} \times (\nabla \times \hat{n})]^2$$

Untwisted chiral LC cell





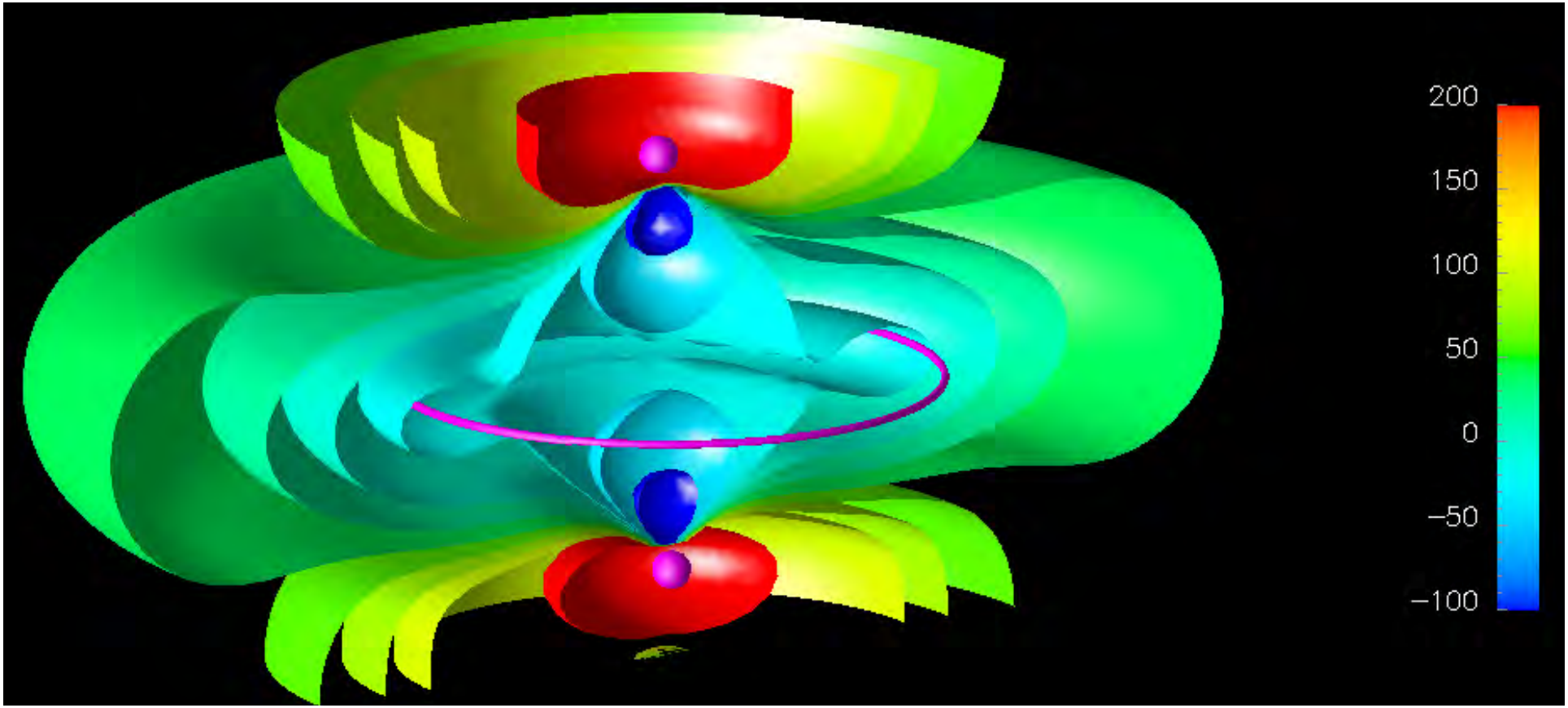
Chiral LC cell with a toron



→Topologically protected: stable unless we induce defects

→Strong energetic barrier > 1000 $K_B T$

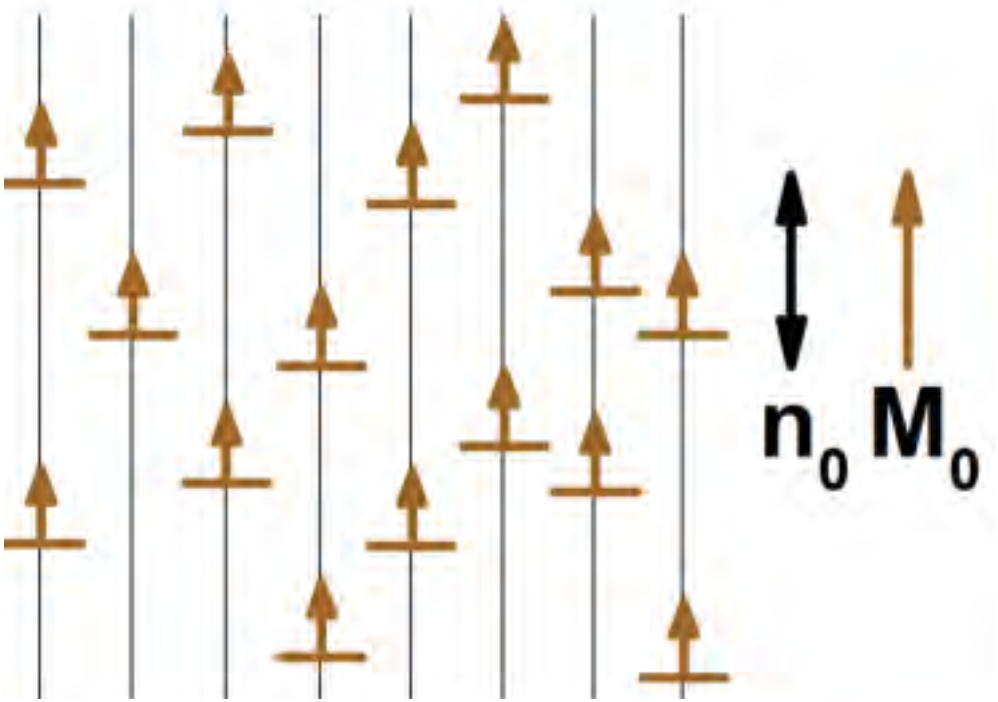
Total free energy density isosurfaces



- The energy is minimized in the twisted part of the structure
- Point defects energetically costly

Ferromagnetic Liquid Crystal Colloids

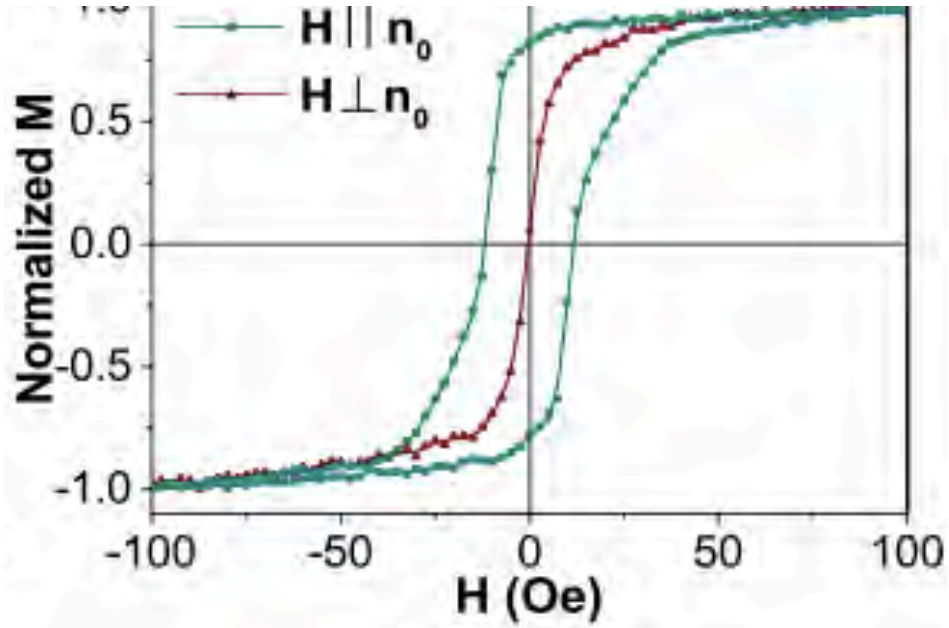
- Platelet-shaped ferromagnetic nanoparticles dispersed in a chiral nematic liquid crystal
- Ferromagnetic nanoplatelets self-align in LC and induce a magnetization field **M(r)** parallel **n(r)**
- Strong sensitivity to weak magnetic field 1-10 G



Mertelj, Alenka, et al. Nature 504, 237, 2013.

Free energy, Numerical Modeling

$$F = F_{elast} + F_{mag} + F_{coupl} + F_{surf}$$



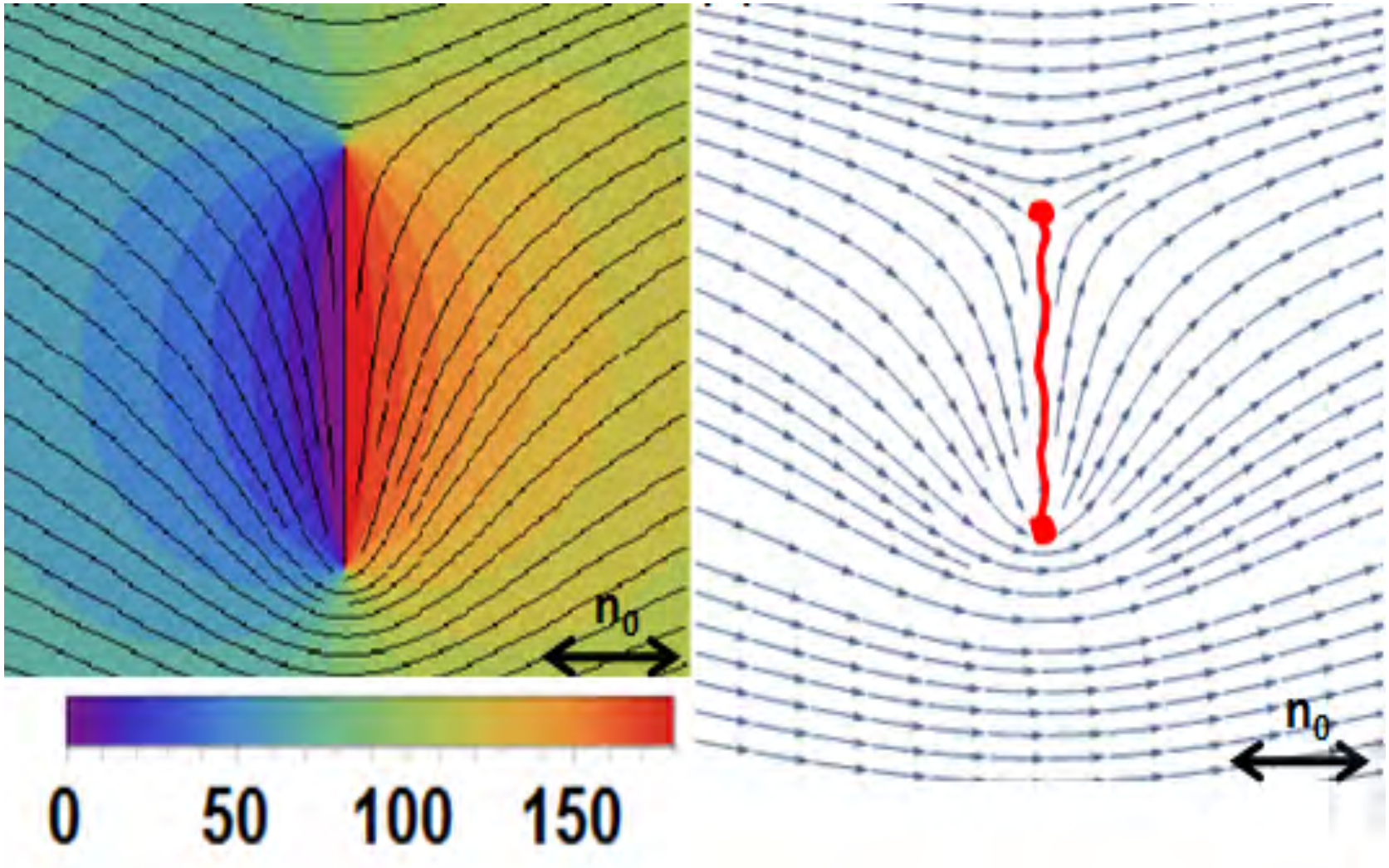
$$F_{\text{elast}} = \frac{1}{2}\int_0^d \left(K_1(\nabla\cdot\mathbf{n})^2 + K_2(\mathbf{n}\cdot(\nabla\times\mathbf{n}))^2 + K_3(\mathbf{n}\times(\nabla\times\mathbf{n}))^2\right)\mathrm{d}x$$

$$F_{mag} = -\int_0^d \mu_0 M\cdot H dx$$

$$F_{\text{coupl}} = -\frac{1}{2}\int_0^d \gamma\mu_0(\mathbf{n}\cdot\mathbf{M})^2\mathrm{d}x$$

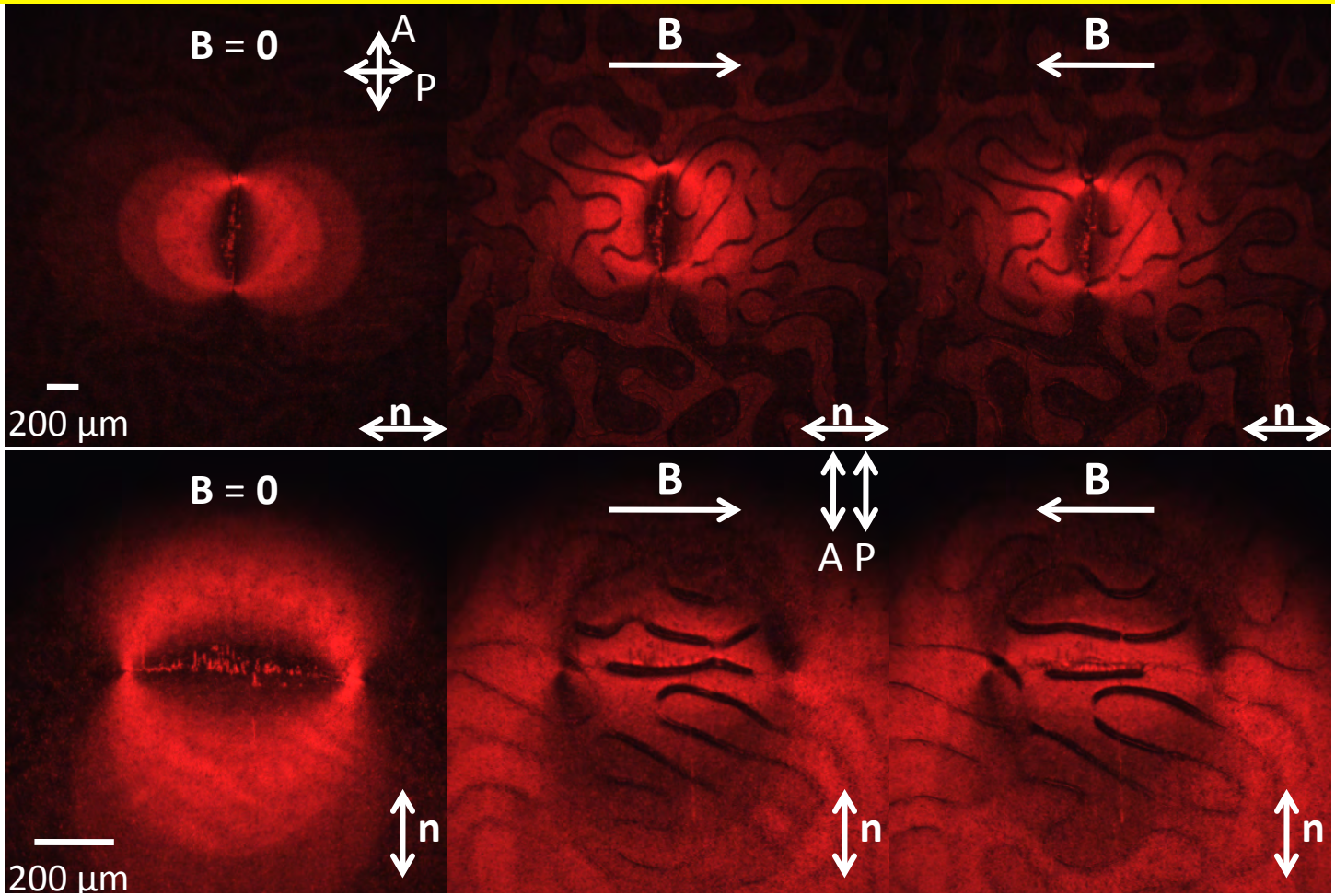
$$F_{\text{surf}} = -\frac{1}{2}W(\mathbf{n}\cdot\mathbf{n}_{\text{s}})^2$$

Optical Patterning of Director & Magnetization Fields



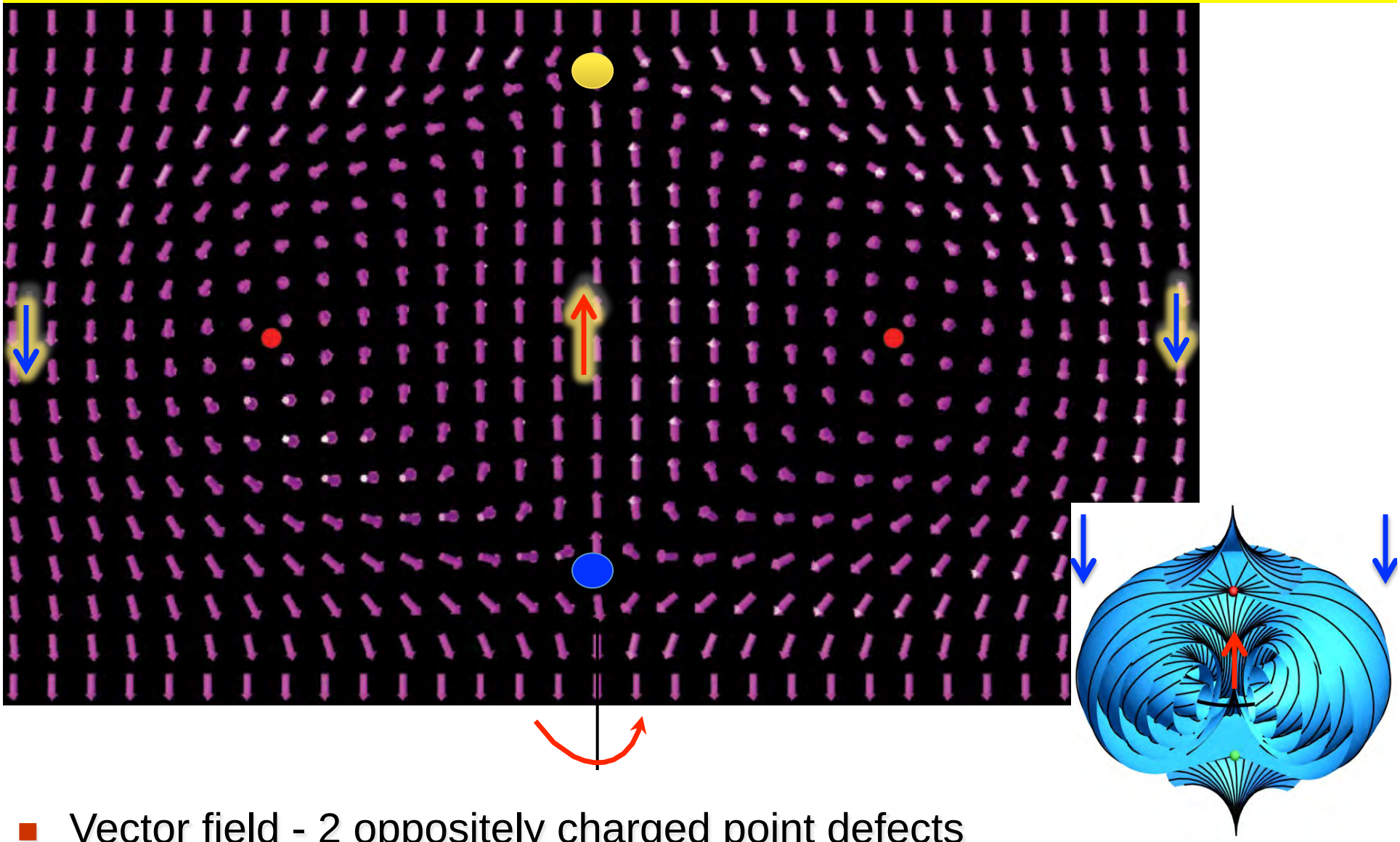
→Isolated half-integer defect lines allowed in the director field but not in **M**(**r**)
→Thus obtain a wall defect in **M**(**r**)

Patterning of defects in director & magnetization



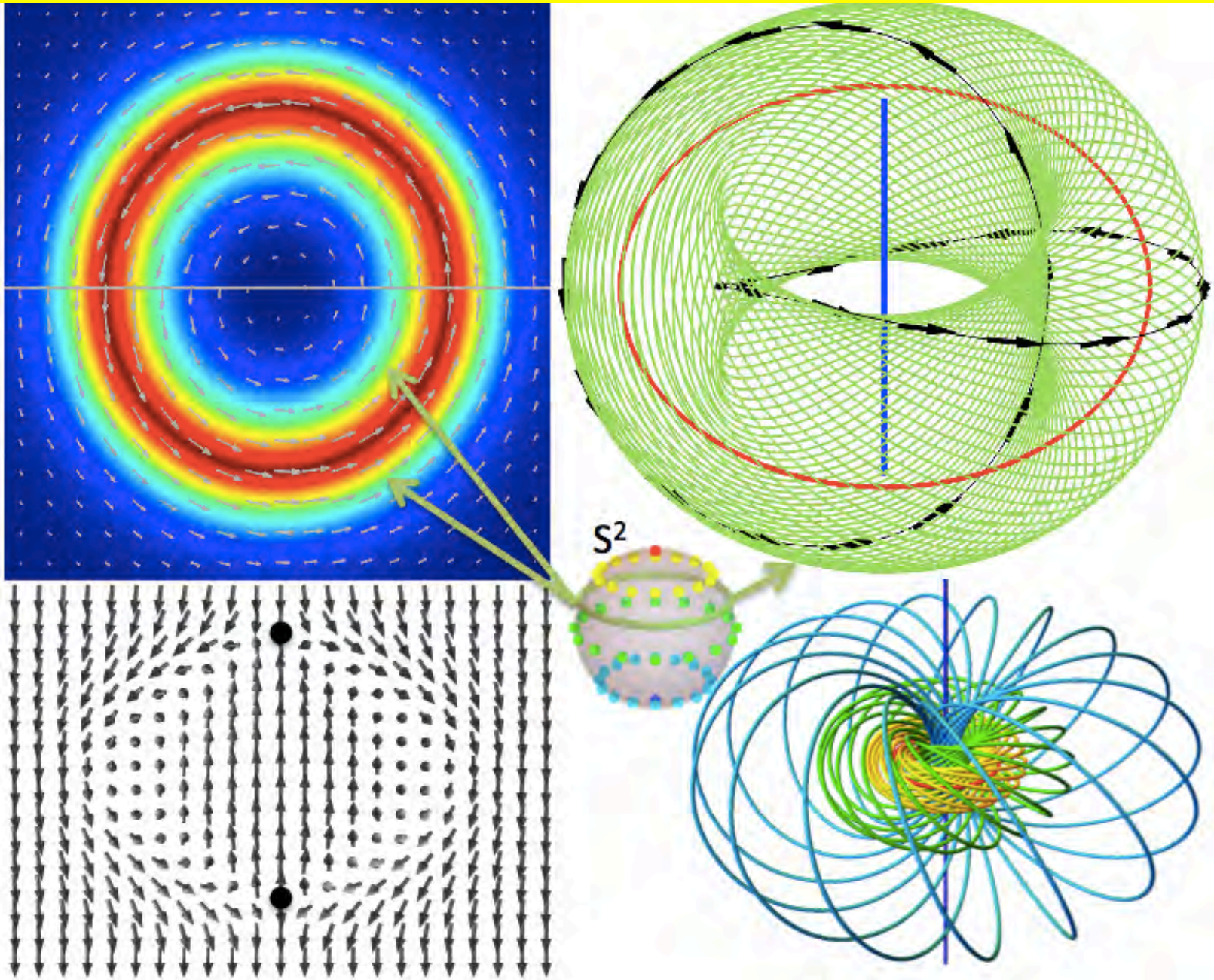
→The wall defect in **M**(**r**) is clearly visible at no applied fields & interacts with domain walls **B** is applied

Toron vs. Hopfion: point defects required

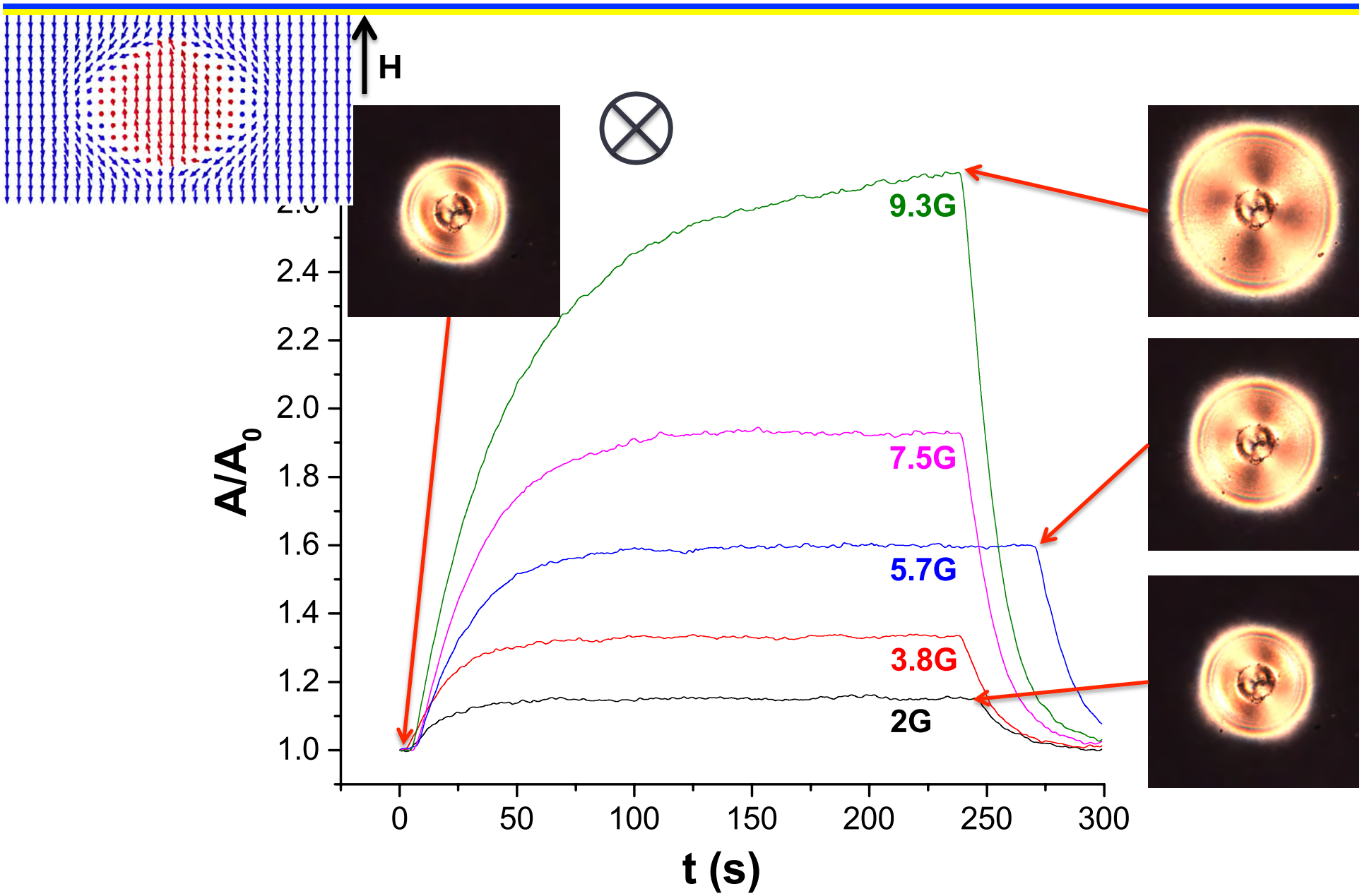


- Vector field - 2 oppositely charged point defects
- Help matching the vector field to the far-field **M**

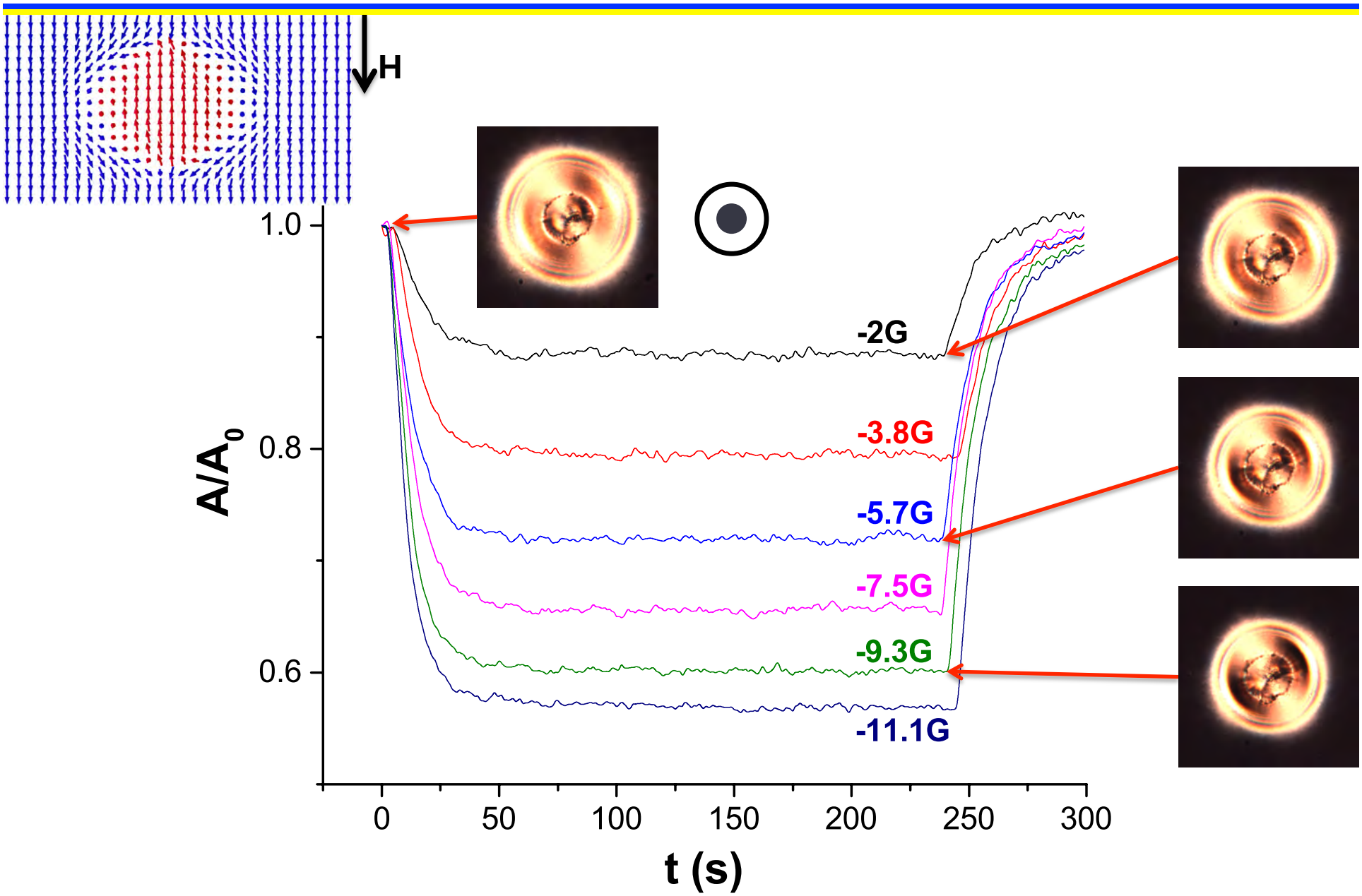
Topology of magnetization field lines M(r)



Toron lateral size vs. field

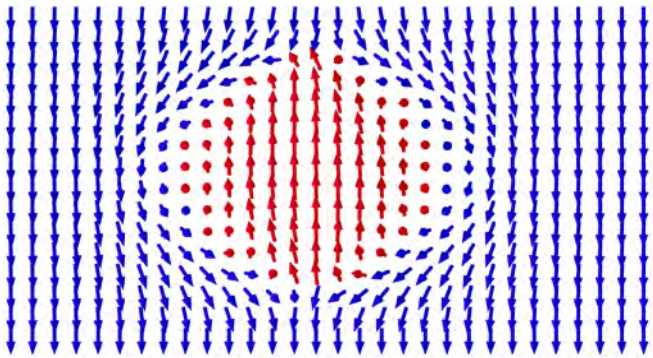


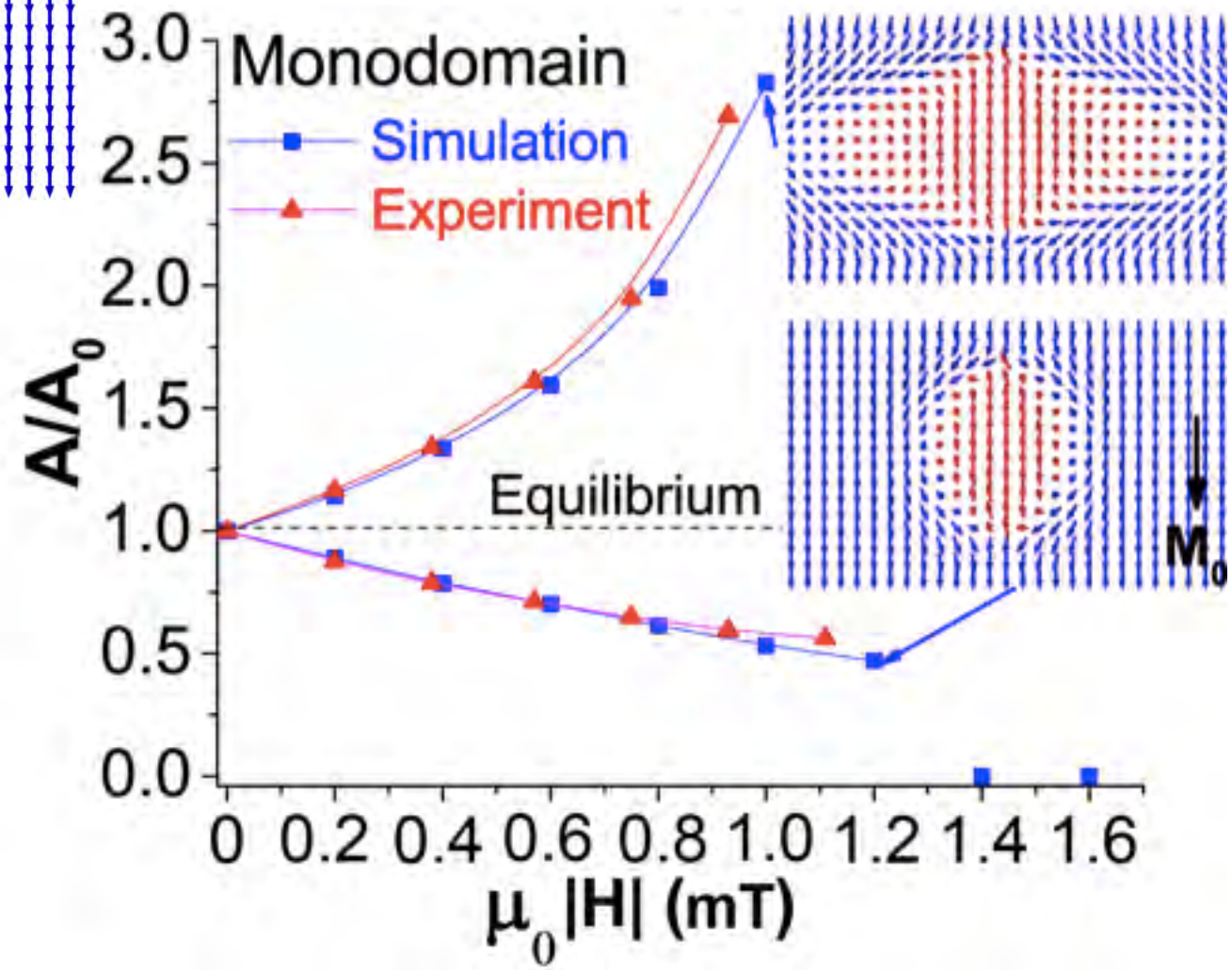
Toron lateral size vs. field



Comp. simulation: toron lateral size vs. field

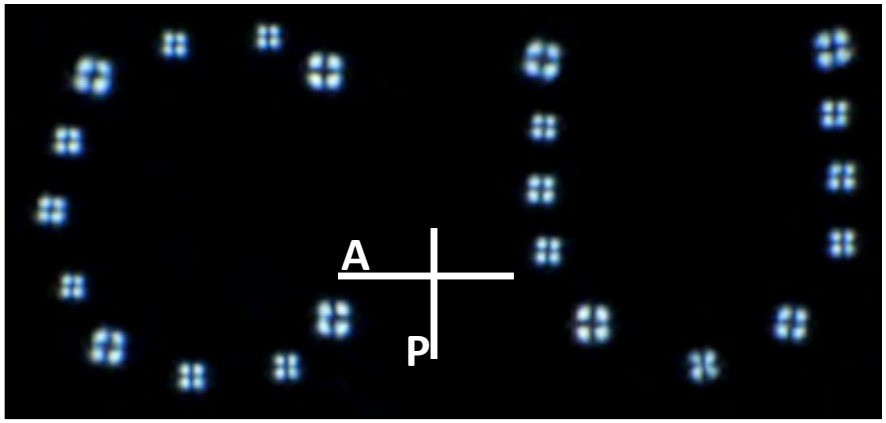
Equilibrium at H=0



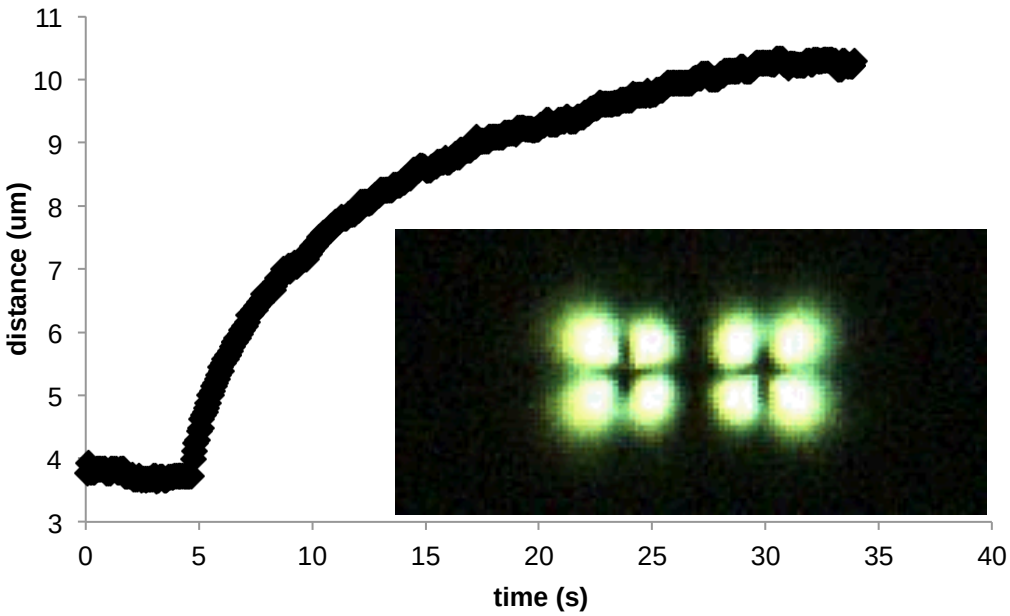


Modes of generation: sticking & mobile

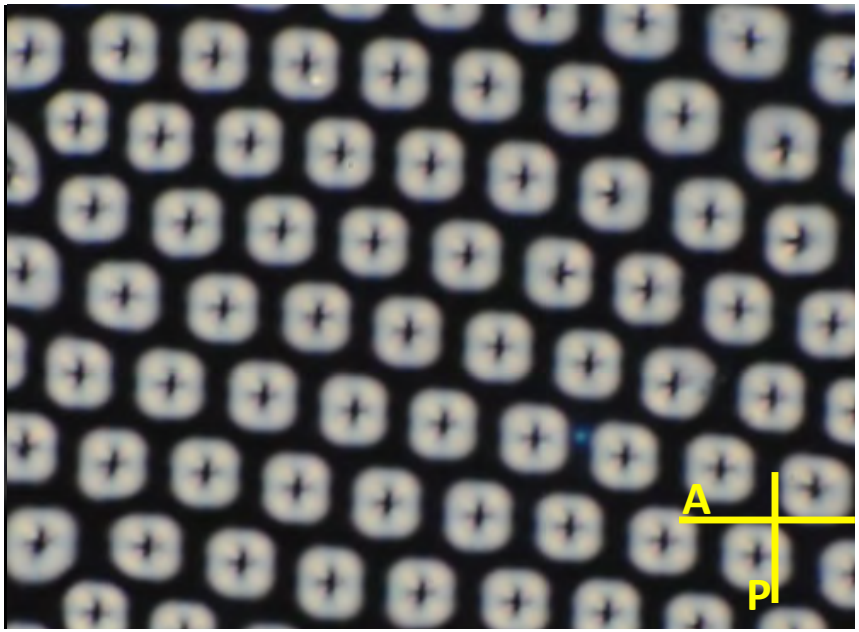
At powers >70mW:
→Pinned where generated;



At powers 30-70mW:
→Repulsion;
→Brownian particle-like motion



Dense arrays & crystallization due to confinement & repulsive interactions

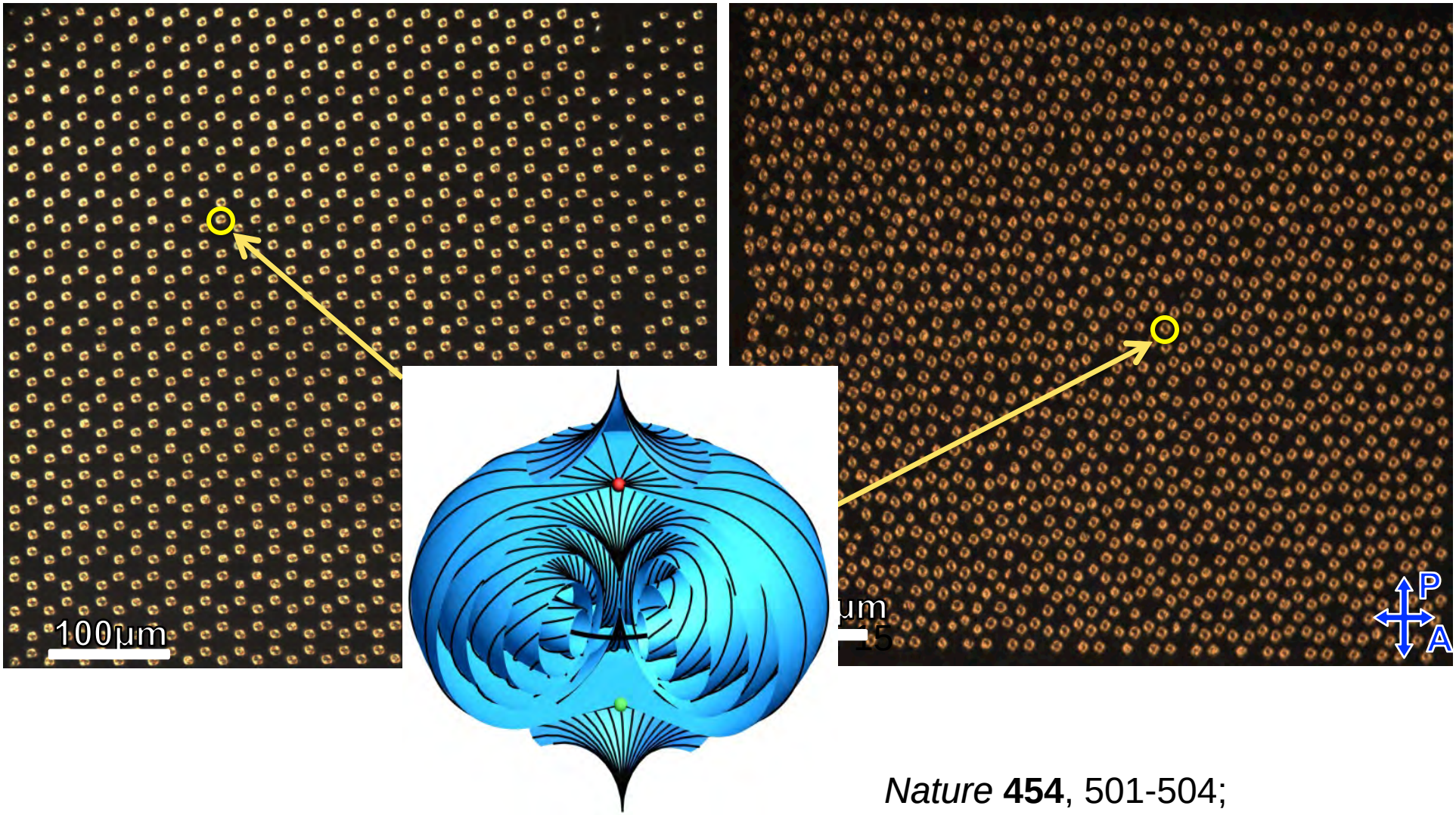


Patterning of defects & control of optical solitons

Periodic lattices & quasicrystal-like structures

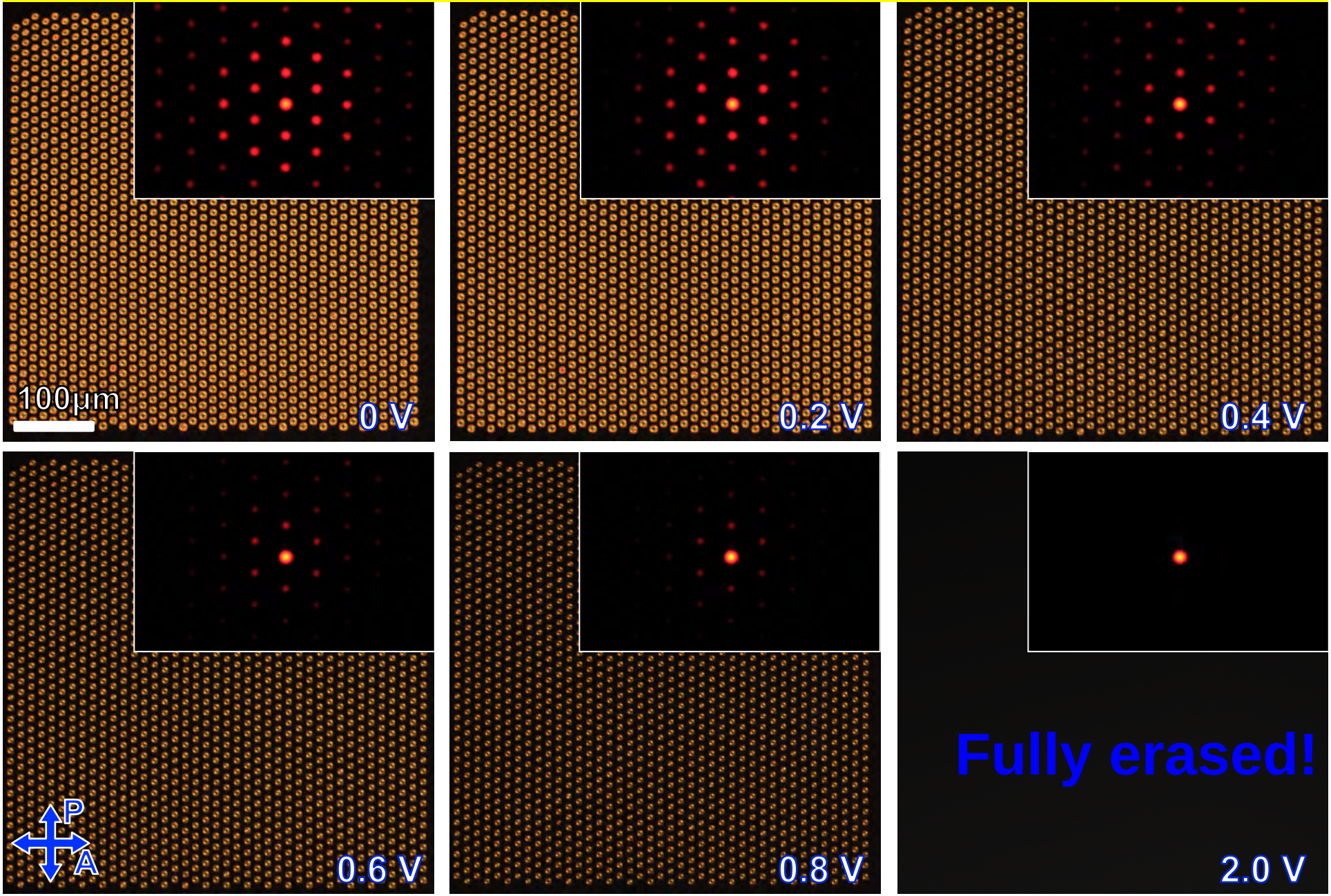
Honeycomb

Quasicrystal-like pattern

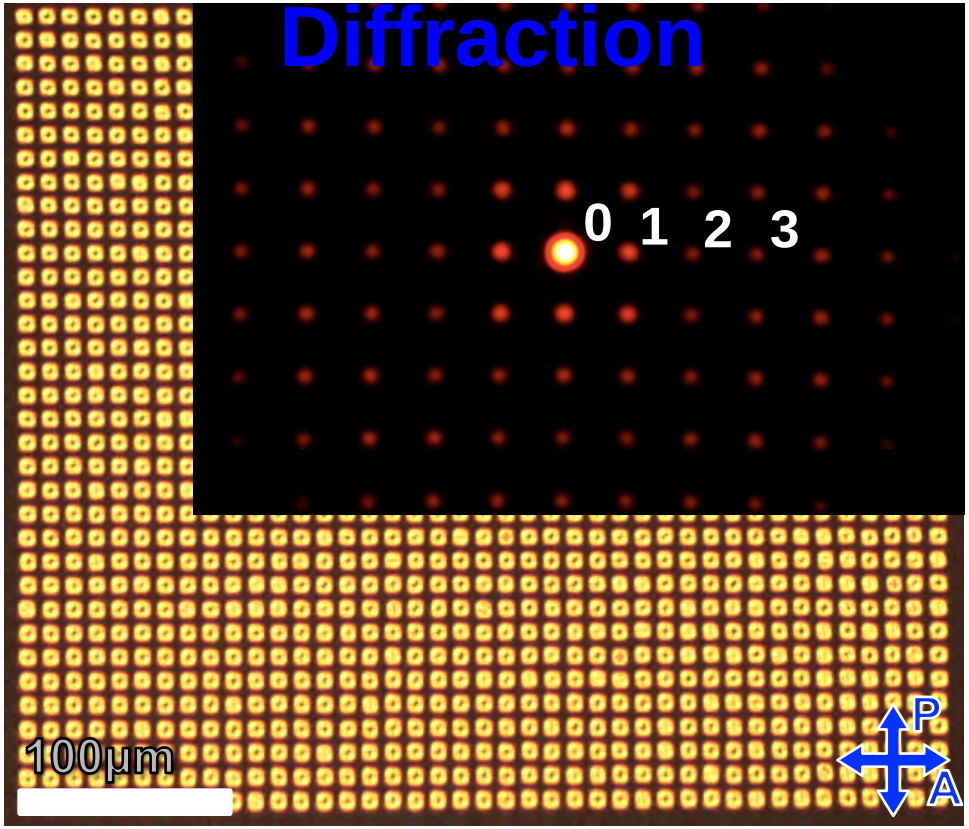


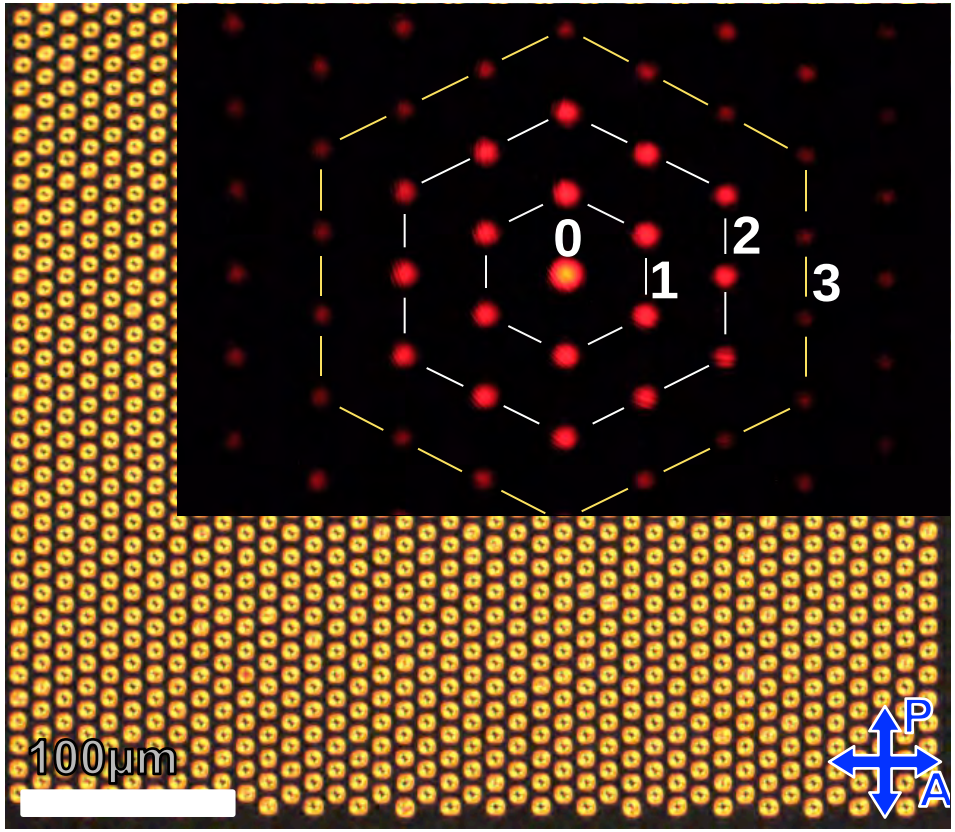
Nature **454**, 501-504;
Ackerman et al, Phys. Rev. E **86**, 021703 (2012)

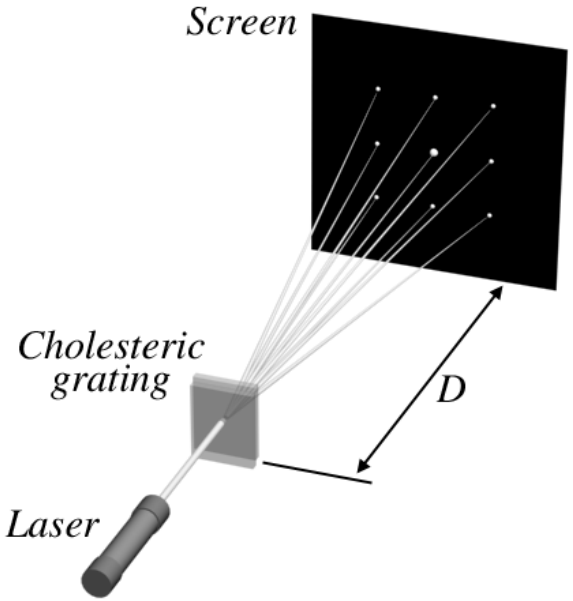
Example of tuning/erasing toron gratings



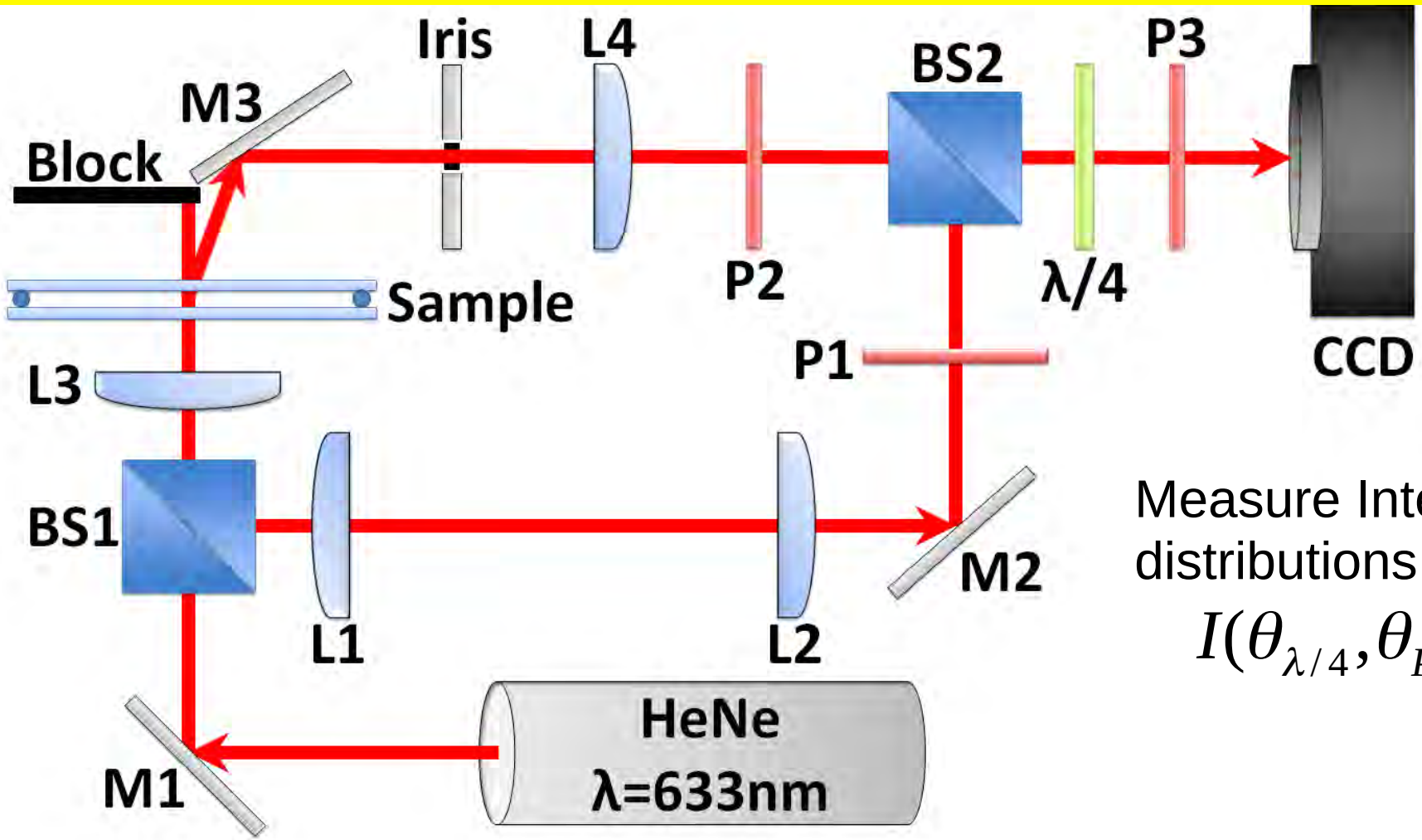
Ackerman, Qi, & Smalyukh, Phys Rev E (2012)


Diffraction
0 1 2 3
100µm
P
A


0
1
2
3
100µm
P
A


Screen
Cholesteric grating
D
Laser

Stokes Polarimetry for Phase Mapping



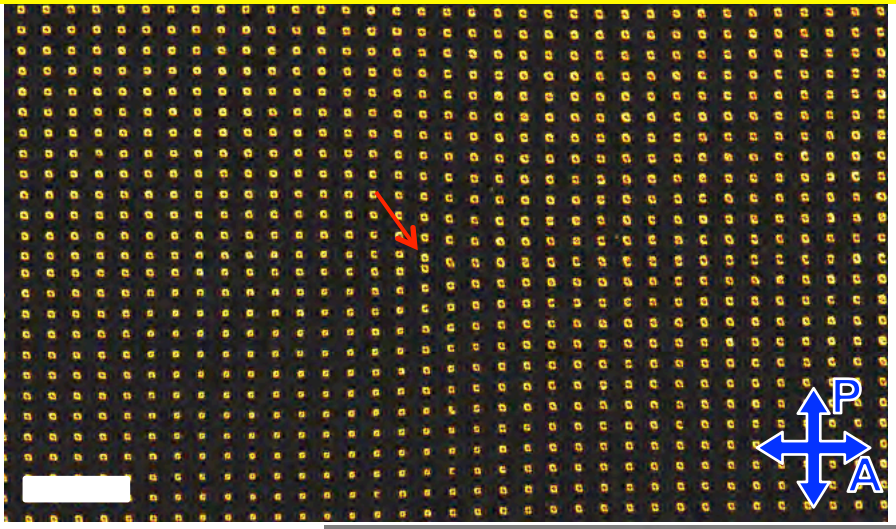
Measure Intensity distributions

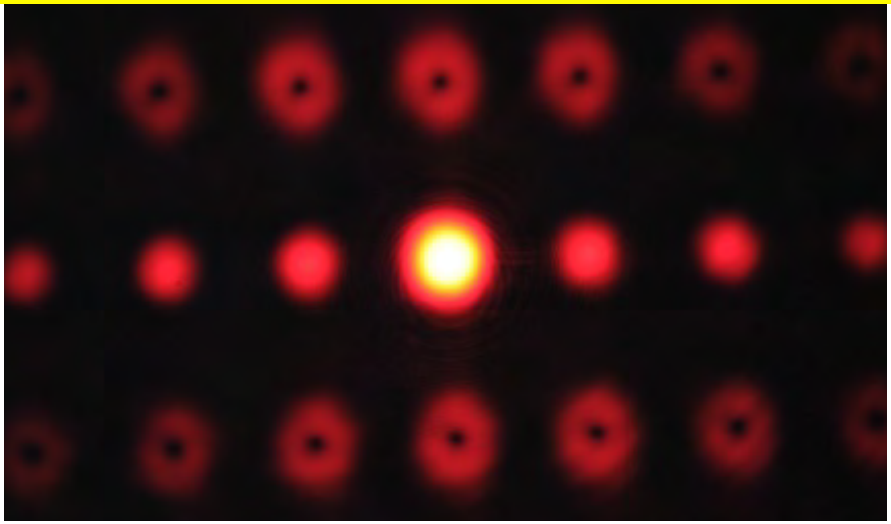
$$I(\theta_{\lambda/4}, \theta_P)$$

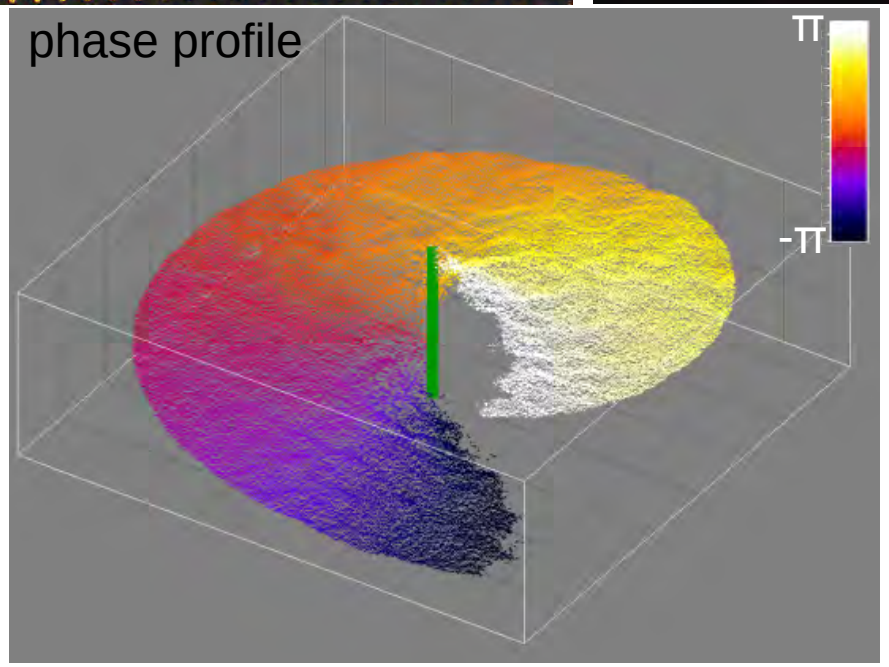
$$S_2 = I(\pi/4, \pi/4) - I(3\pi/4, 3\pi/4) \qquad S_3 = I(\pi/4, 0) - I(3\pi/4, 0)$$

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

Phase singularities generated by toron arrays

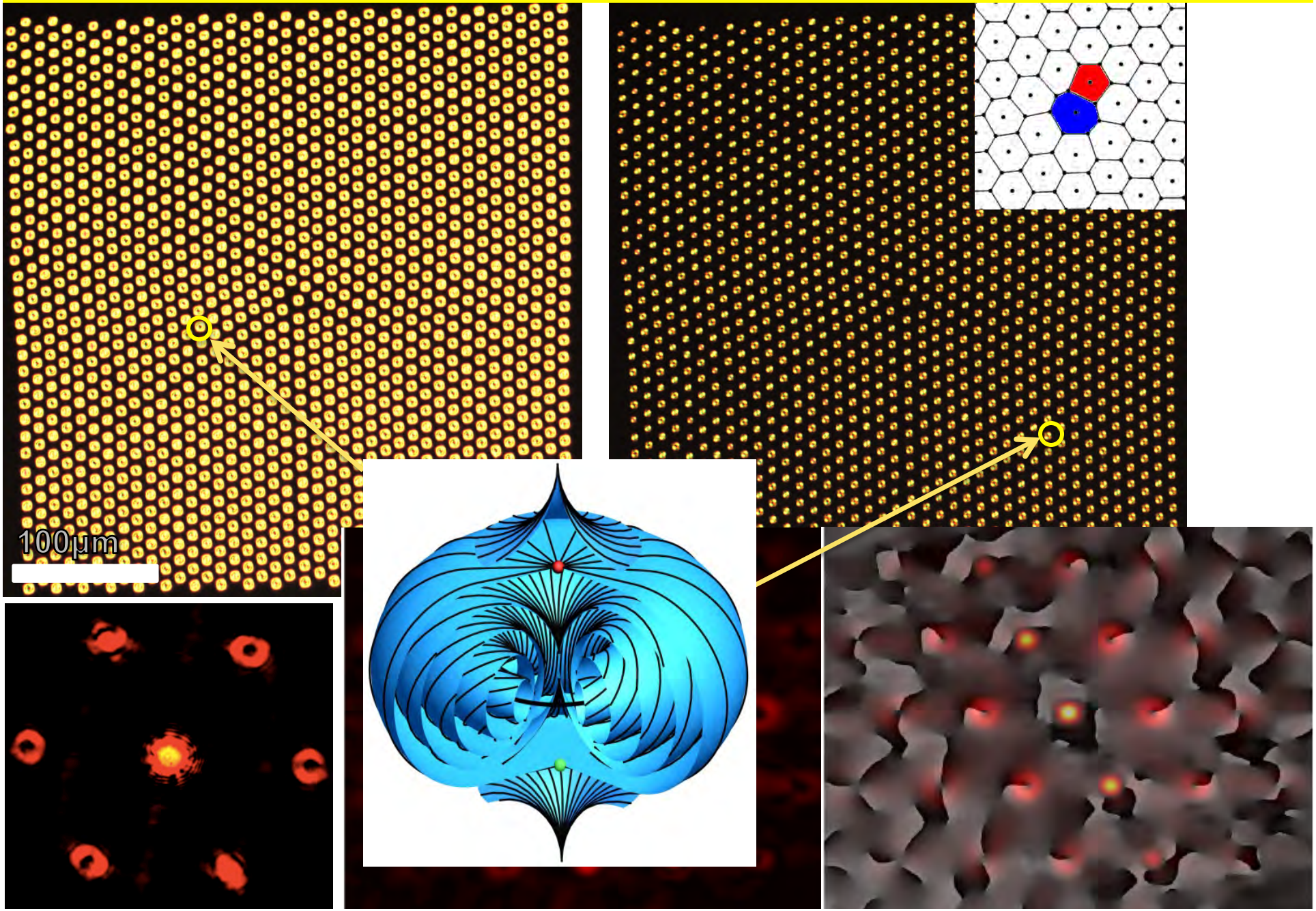






P. J. Ackerman, Z. Qi, Y. Lin, C. W. Twombly, M. J. Laviada, Y. Lansac & I. I. Smalyukh, *Scientific Reports* **2**, 414 (2012)

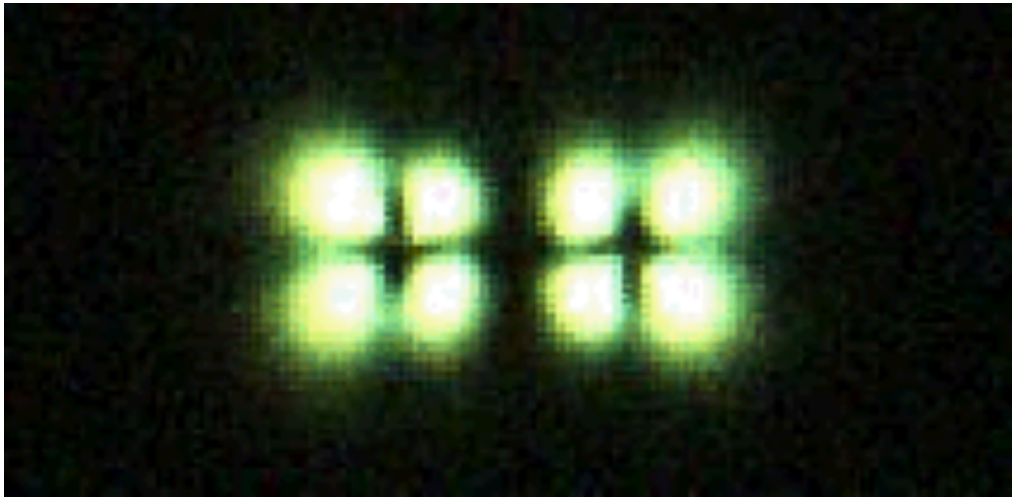
5-7 defects in toron arrays & phase singularities

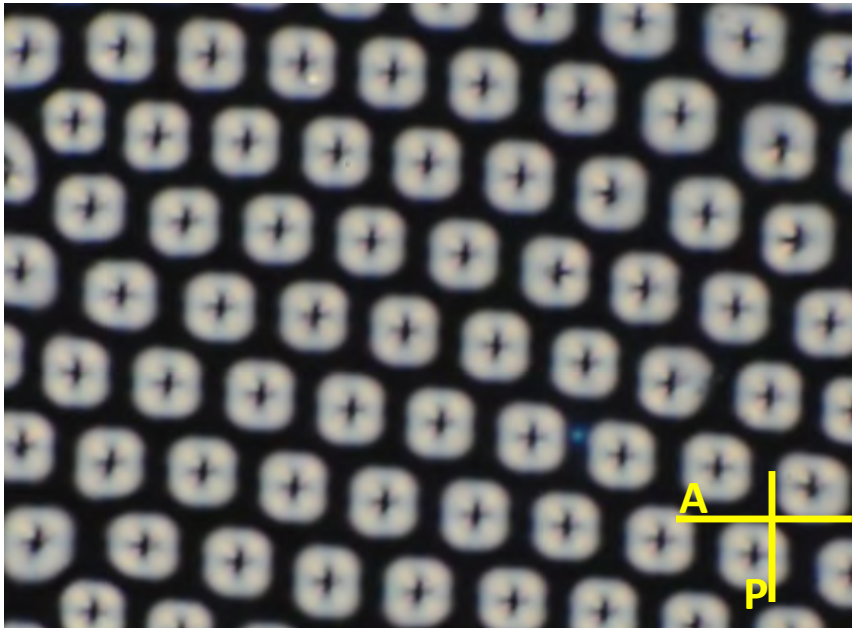


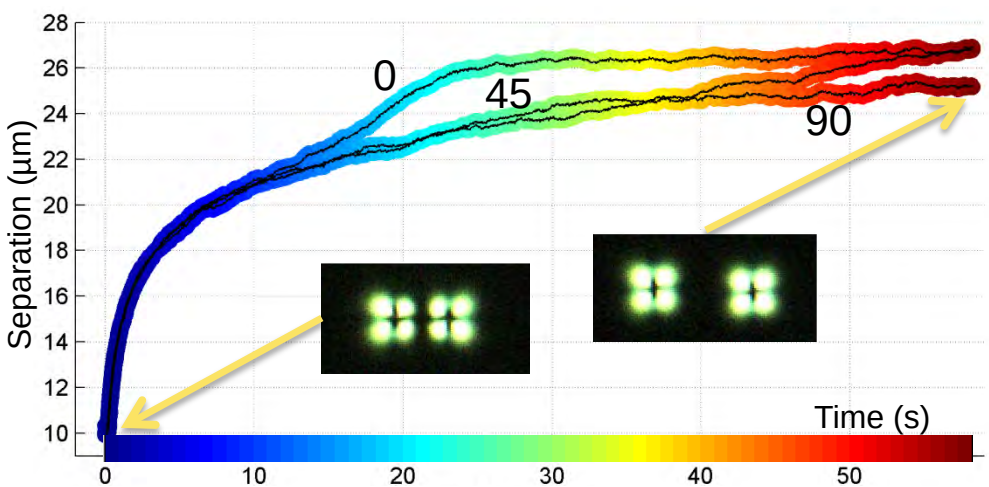
Self-assembly & non-equilibrium dynamics of knotted 3D solitons

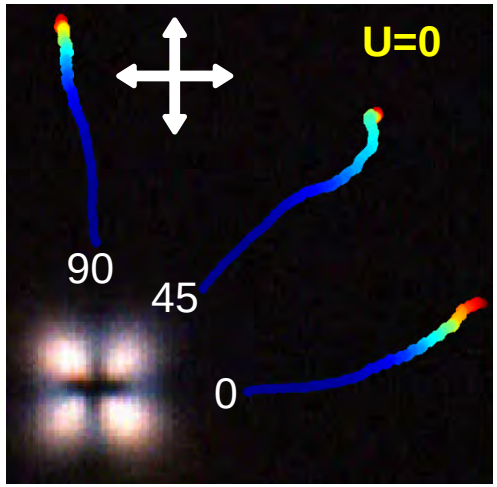
Pair interactions & 2D crystallization

Dense arrays & crystallization





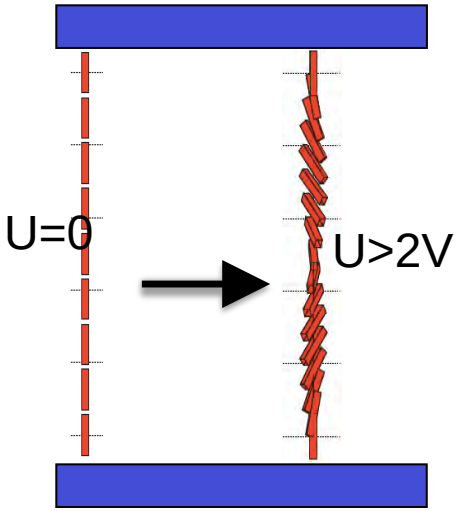




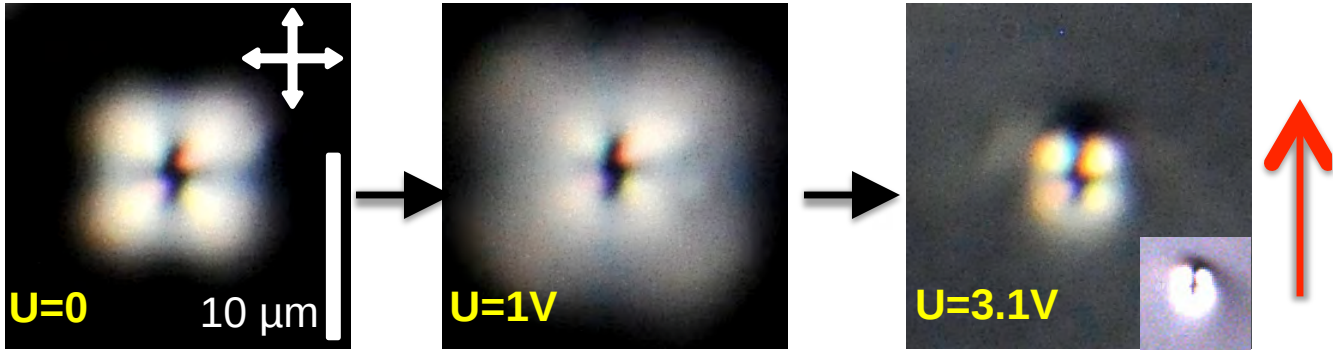
P. J. Ackerman, J. van de Lagemaat, and I. I. Smalyukh. *NATURE COMMUNICATIONS* 6, 6012 (2015)

Voltage-reconfigurable structure

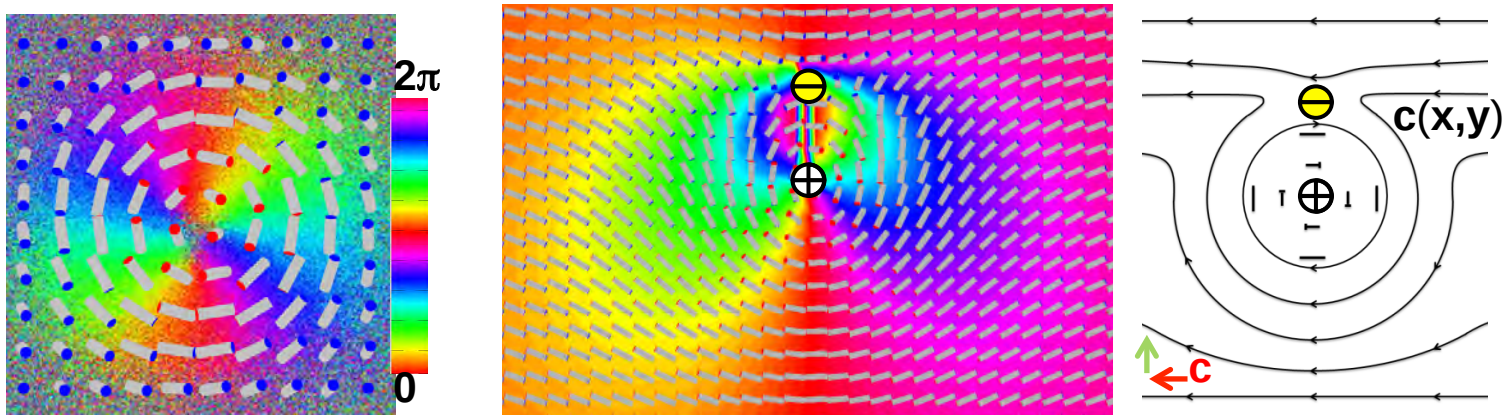
vertical cross-section



→Increasing voltage

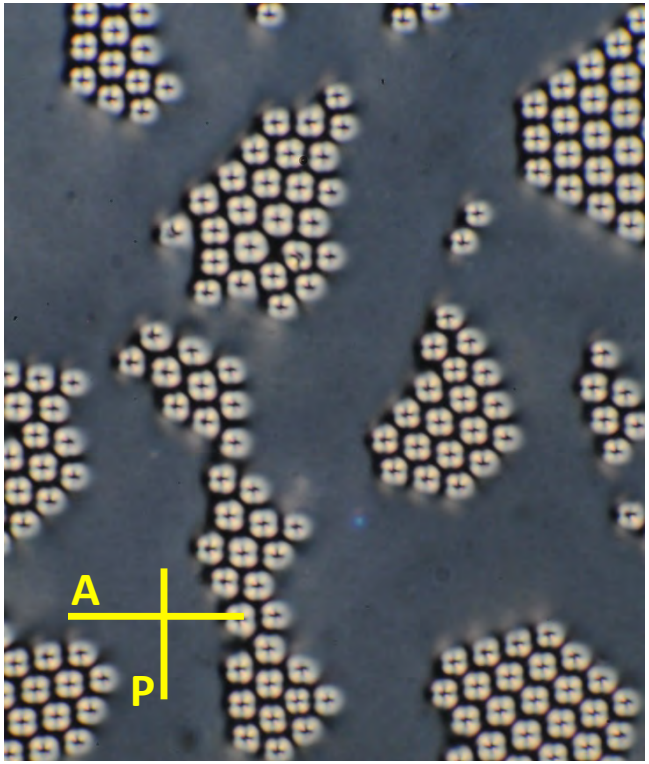


→Structural transformation with umbilical defect formation

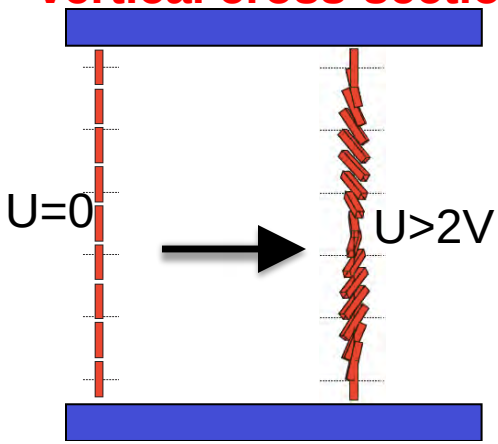


Voltage-reconfigurable self-assembly

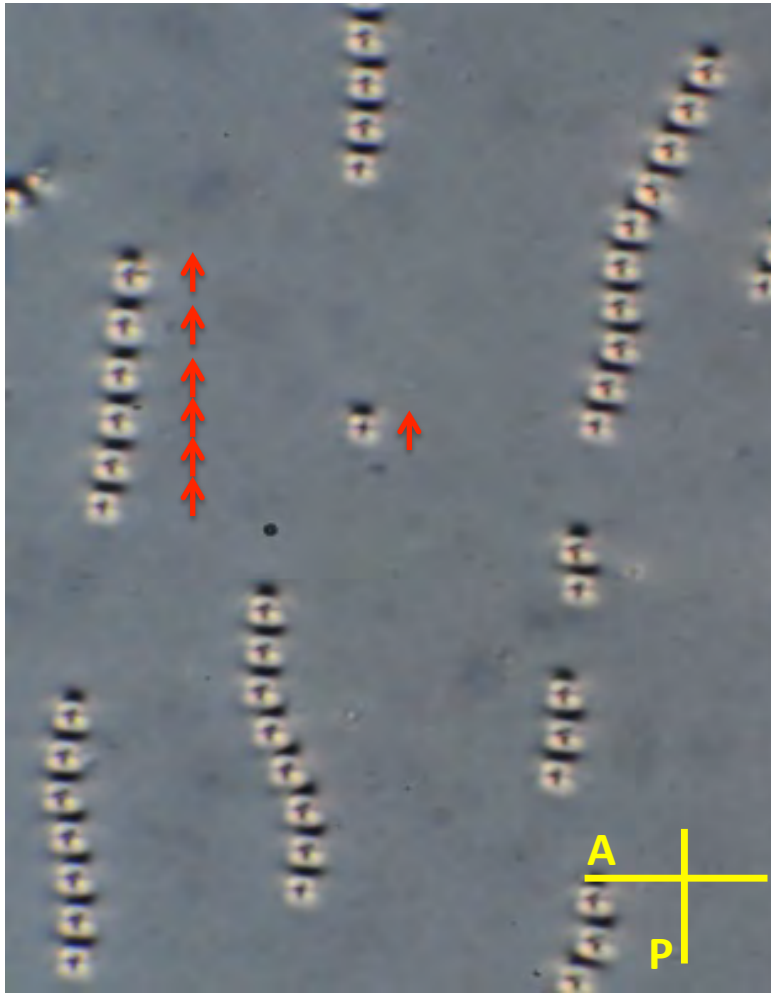
→Attractive isotropic @ 2-3V

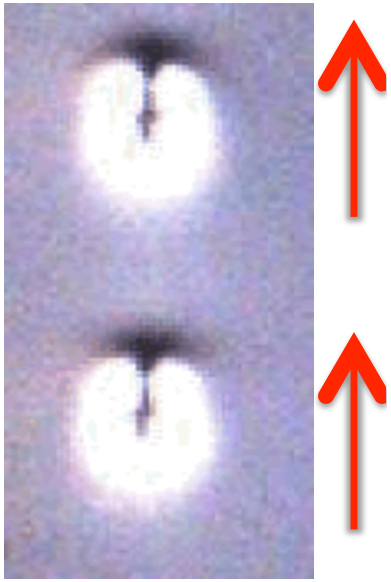


vertical cross-section



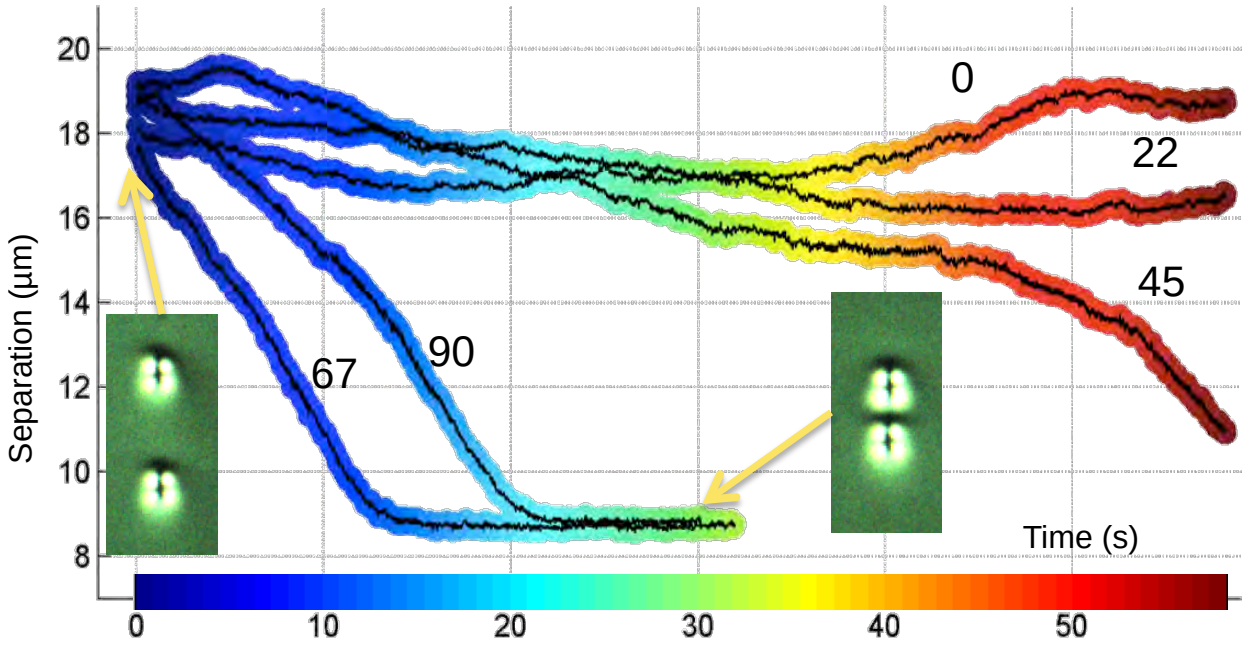
→Attractive anisotropic dipolar-like @ 3-5V

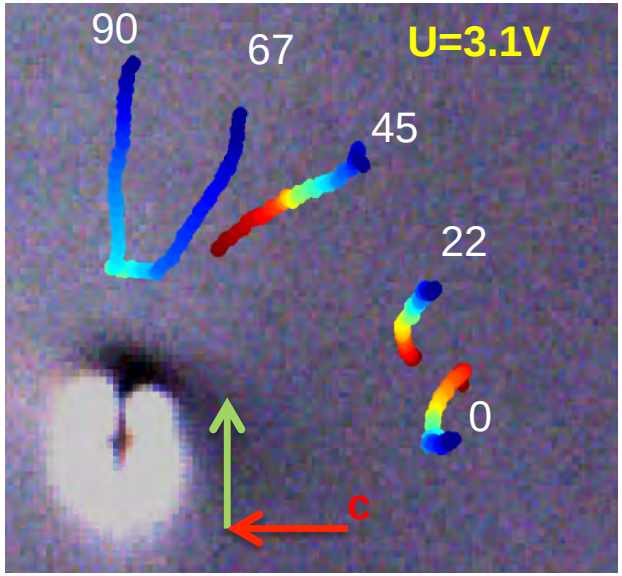




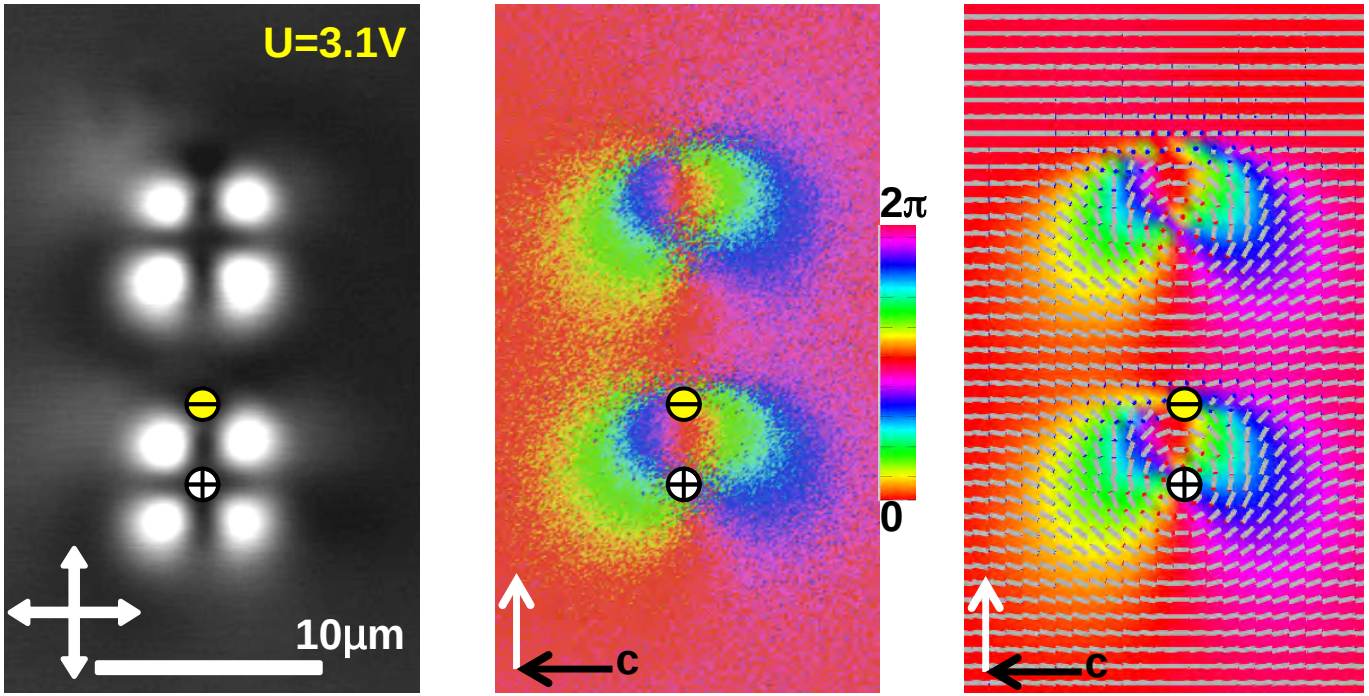
→Knowing topology can control self-assembly by U

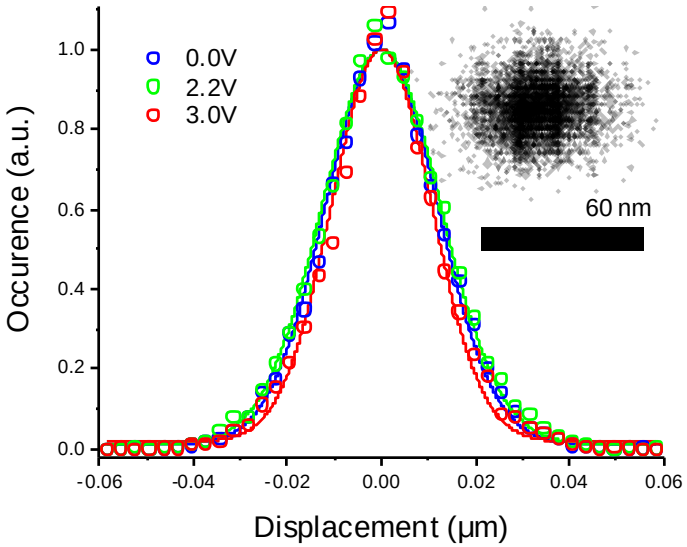
Field-controlled pair interactions

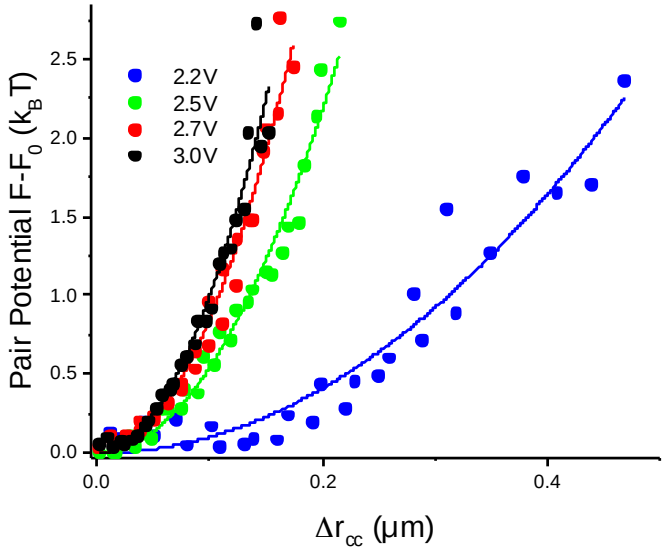




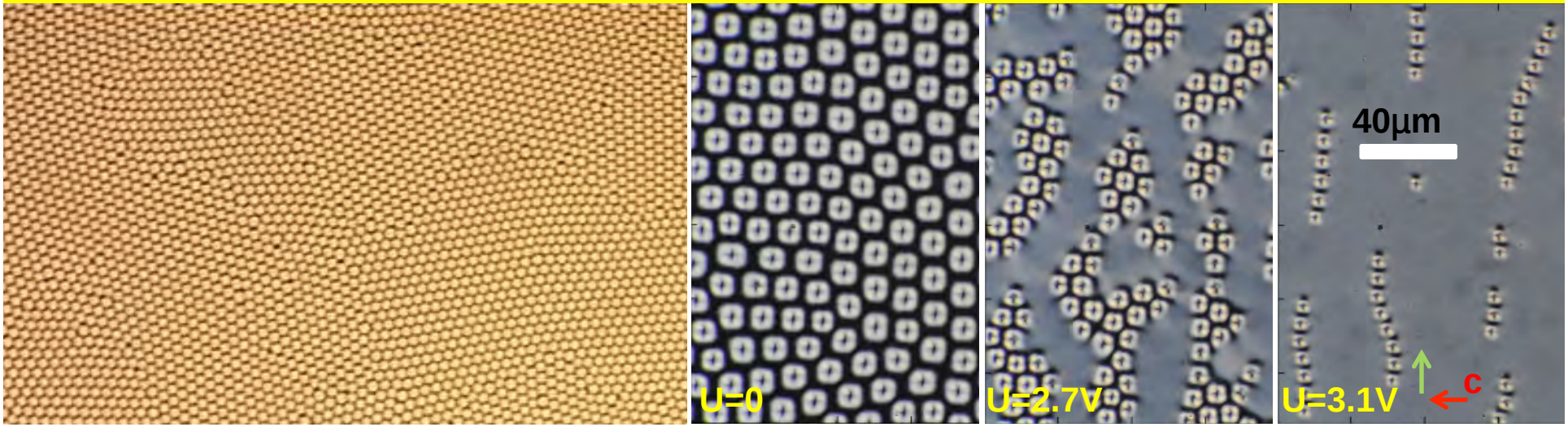
Field-controlled pair interactions

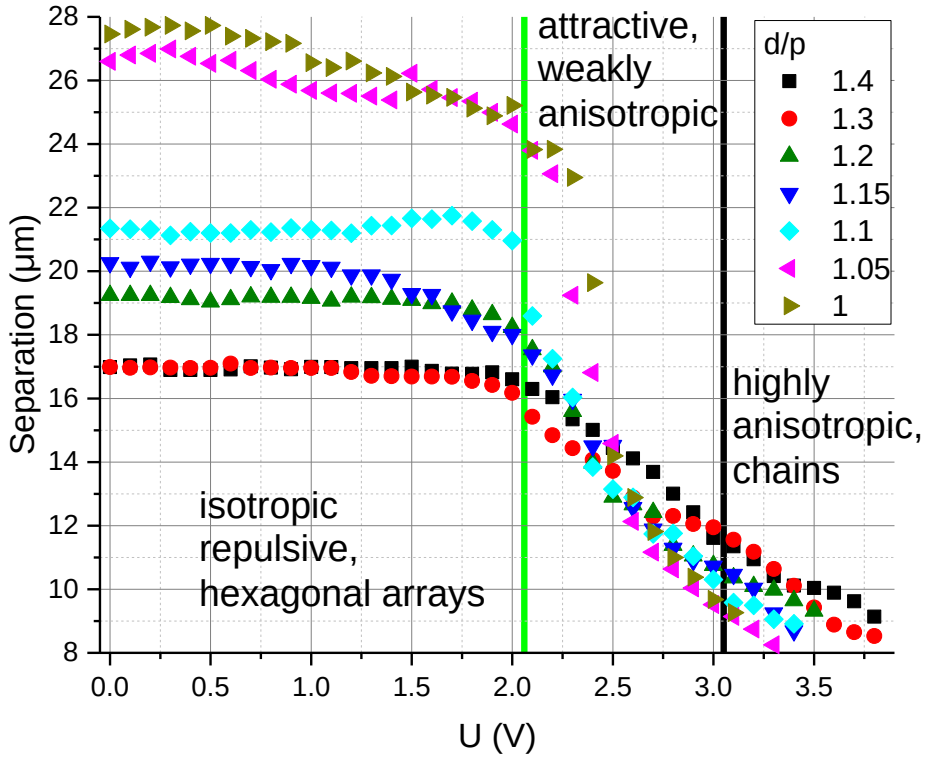


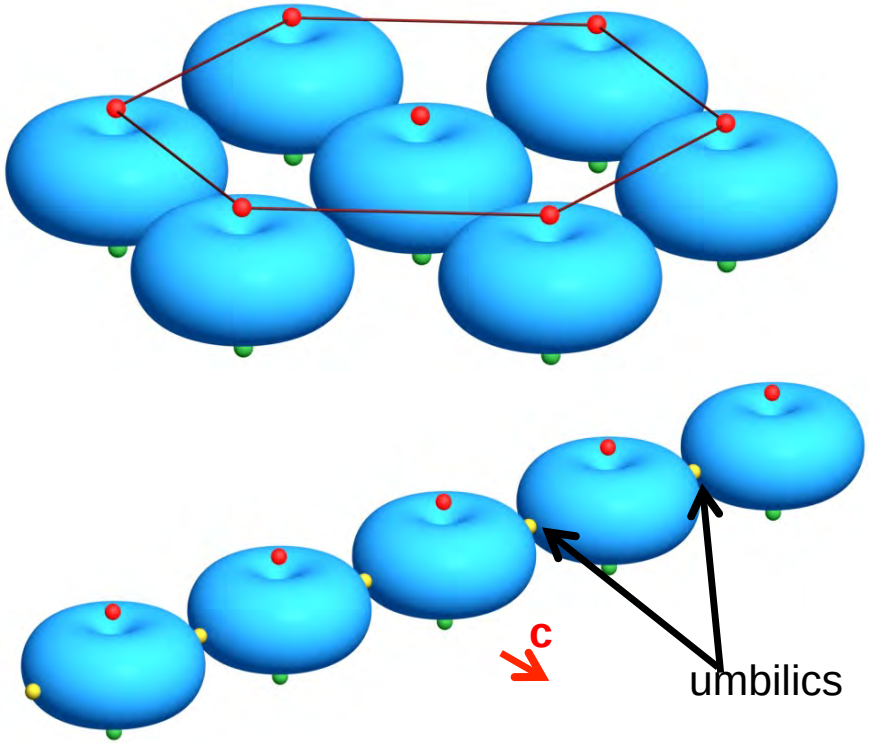




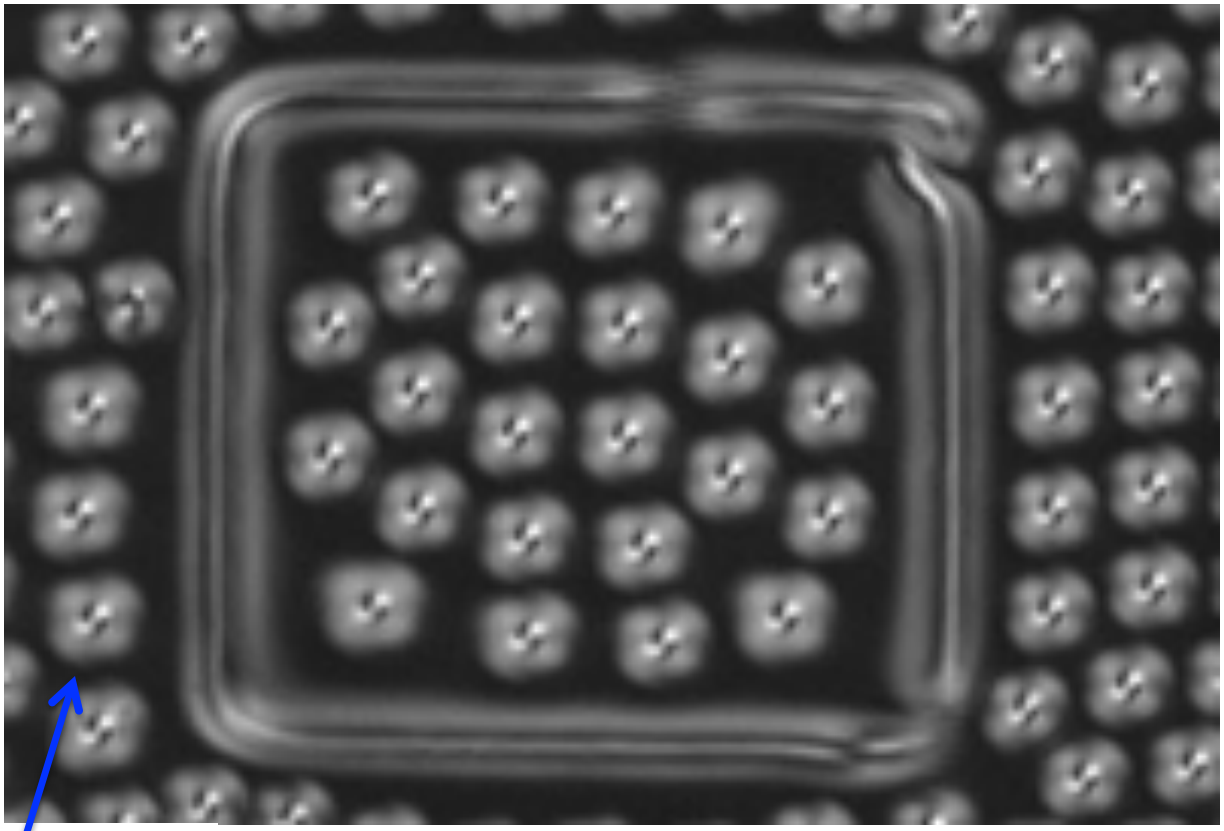
Voltage-reconfigurable self-assembly

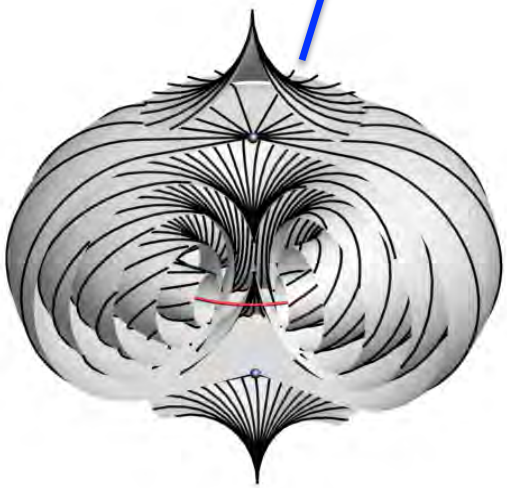






Light-induced dynamics of topological solitons





→Non-equilibrium dynamics of 2D & 3D solitons

Conclusions

→Soft Matter provides an experimental toolkit of exploring topology;

→Interplay of defects in light & matter

→Topology – the basis for controlling self-assembly

Collaborators

- M. Tasinkevych, MPI, Germany
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- J. Yeomans, Oxford
- S. Zumer & M. Ravnik, Ljubljana, Slovenia