School of Modern Optics

6 May 2013, Puebla, Mexico

Lecture 1

Liquid crystals under optical fields

Etienne Brasselet

Singular Optics & Liquid Crystals group

www.loma.cnrs.fr/spip.php?rubrique331

Laboratoire Ondes et Matières d'Aquitaine
CNRS, Université Bordeaux 1, France

Outline

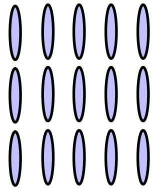
1. **A (very) short introduction to liquid crystals**
2. Dielectric and optical torques
3. Orientational optical nonlinearities
4. Role of the polarization state of light
5. Light-induced nonlinear rotations

“Mesomorphic states of matter” (G. Friedel, 1922)

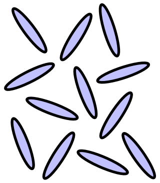
Cristal

Positional order

Orientational order



Liquid



Positional disorder

Orientational disorder

Intermediate but distinct states of matter

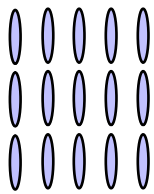
G. Friedel

“Mesomorphic states of matter” (G. Friedel, 1922)

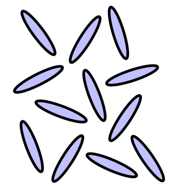
Cristal

Positional order

Orientational order



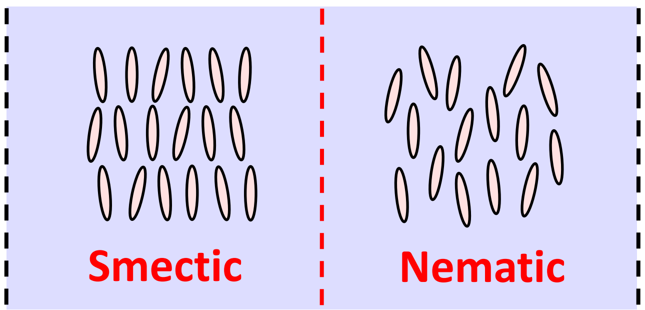
Liquid



Positional disorder

Orientational disorder

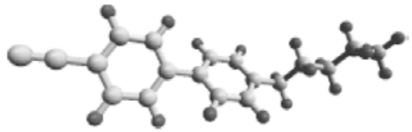
Example



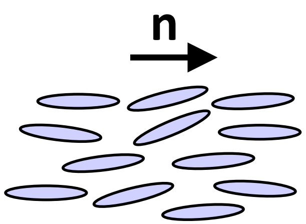
Liquid crystals

G. Friedel

Main characteristics of nematic liquid crystals



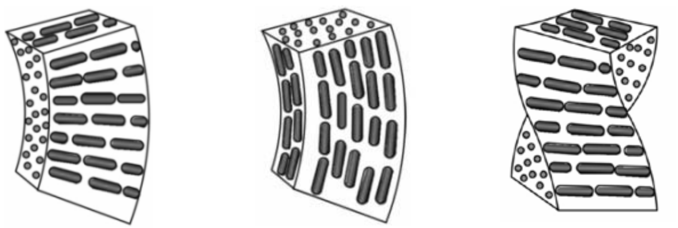
Anisotropy at microscopic scale



Director

$\mathbf{n} \leftrightarrow -\mathbf{n}$

Local average molecular orientation



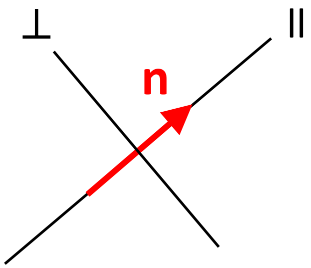
Splay

Bend

Twist

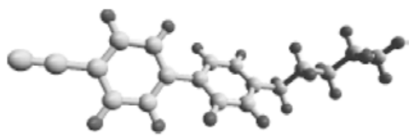
Elastic anisotropy

(Frank constants K_1 , K_2 , K_3)

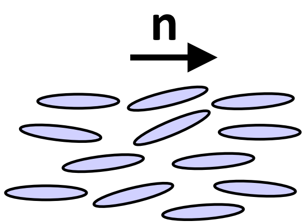


Dielectric anisotropy

Main characteristics of nematic liquid crystals



Anisotropy at microscopic scale



Director

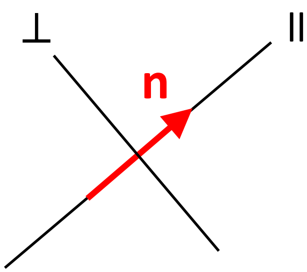
n $\leftrightarrow$ -n

Local average molecular orientation

Splay **Bend** **Twist**

Elastic anisotropy

(Frank constants K_1 , K_2 , K_3)

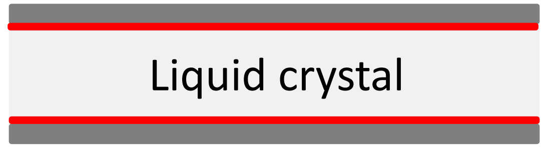


Dielectric anisotropy

$$\overleftrightarrow{\varepsilon} = \varepsilon_0 \begin{pmatrix} \varepsilon_\perp & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_{//} \end{pmatrix}$$

A nematic sample in the lab

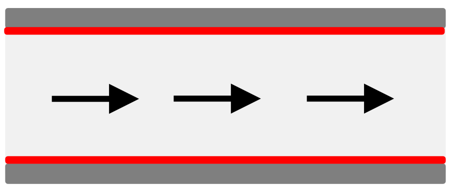
Alignment layers



Glass substrates

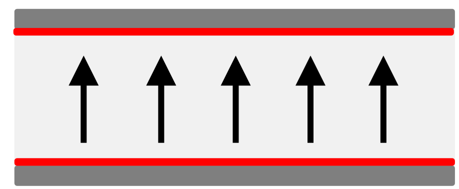
Parallel alignment

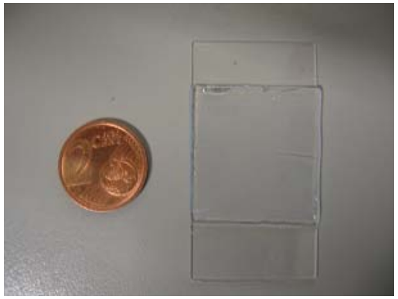
n



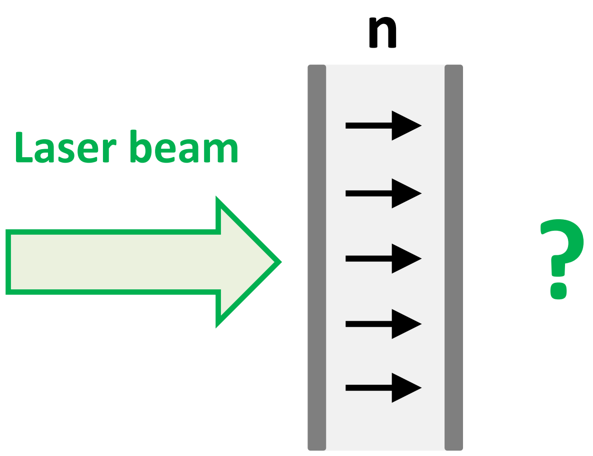
Perpendicular alignment

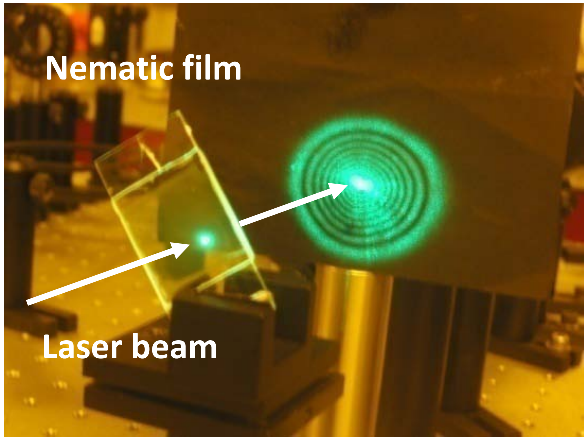
n





Typical thickness : 10 - 100µm





Beam structure is strongly modified

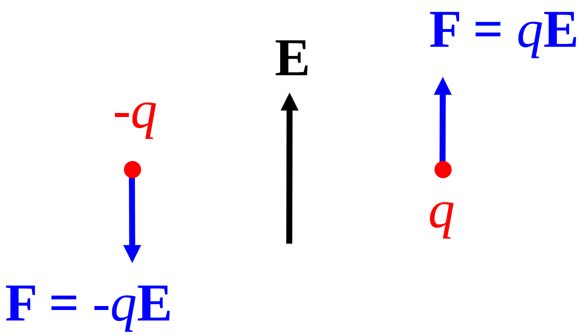
Equation to solve

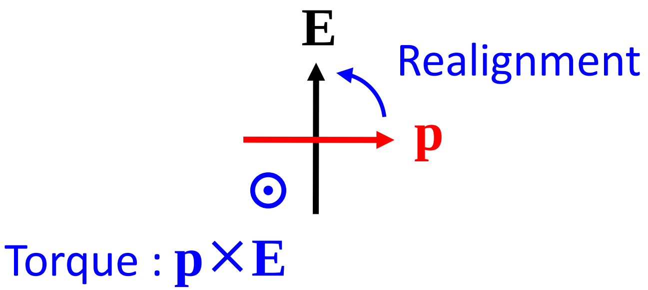
Light + dielectric anisotropy + elasticity = ???

Outline

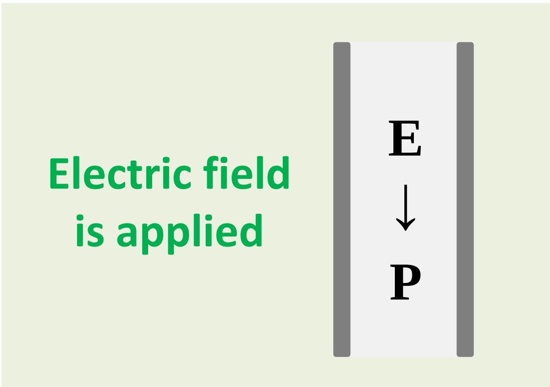
1. A (very) short introduction to liquid crystals
2. **Dielectric and optical torques**
3. Orientational optical nonlinearities
4. Role of the polarization state of light
5. Light-induced nonlinear rotations

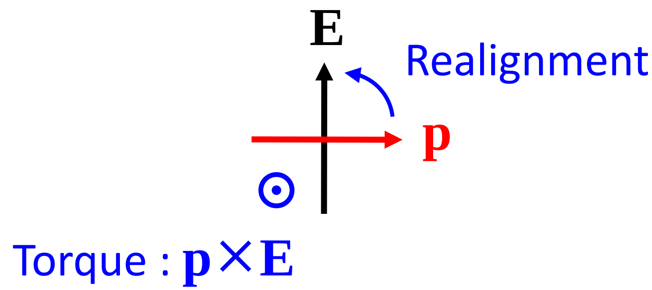
Dielectric torque : fundamentals





Dielectric torque : fundamentals





$$\mathbf{\Gamma} = \mathbf{P} \times \mathbf{E}$$

Dielectric torque per unit volume

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$$

$$\Downarrow$$

$$\boxed{\mathbf{\Gamma} = \mathbf{D} \times \mathbf{E}}$$

$$\mathbf{D} = \overleftrightarrow{\varepsilon} \cdot \mathbf{E}$$

Macroscopic anisotropy required

$$\boldsymbol{\varepsilon}_a = \boldsymbol{\varepsilon}_\parallel - \boldsymbol{\varepsilon}_\perp$$

Dielectric torque on liquid crystals

$$\boxed{\boldsymbol{\Gamma} = (\overleftrightarrow{\varepsilon} \cdot \mathbf{E}) \times \mathbf{E}}$$

(1) Expressions in the local basis of the director "n"

$$\overleftrightarrow{\varepsilon} = \varepsilon_0 \begin{pmatrix} \varepsilon_\perp & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_{//} \end{pmatrix} \qquad \mathbf{E} = \mathbf{E}_\perp + \mathbf{E}_{//}$$

(2) Derivation

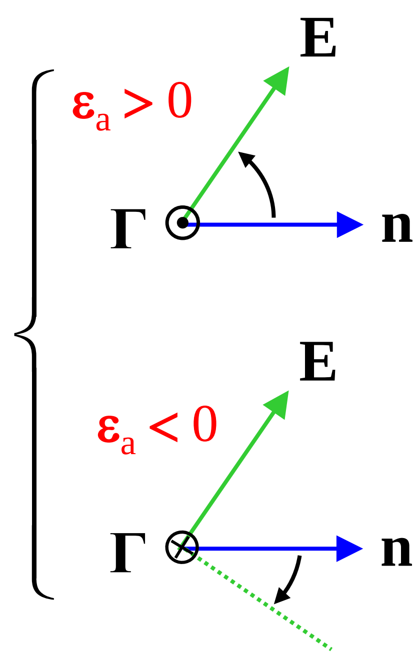
$$\boldsymbol{\Gamma} = \varepsilon_0 (\varepsilon_\perp \mathbf{E}_\perp + \varepsilon_{//} \mathbf{E}_{//}) \times (\mathbf{E}_\perp + \mathbf{E}_{//})$$

$$= \varepsilon_0 (\varepsilon_{//} - \varepsilon_\perp) \mathbf{E}_{//} \times \mathbf{E}_\perp$$

$$= \varepsilon_0 \varepsilon_a (\mathbf{n} \cdot \mathbf{E}) \mathbf{n} \times \mathbf{E}_\perp$$

$$\boxed{\boldsymbol{\Gamma} = \varepsilon_0 \varepsilon_a (\mathbf{n} \cdot \mathbf{E}) (\mathbf{n} \times \mathbf{E})}$$

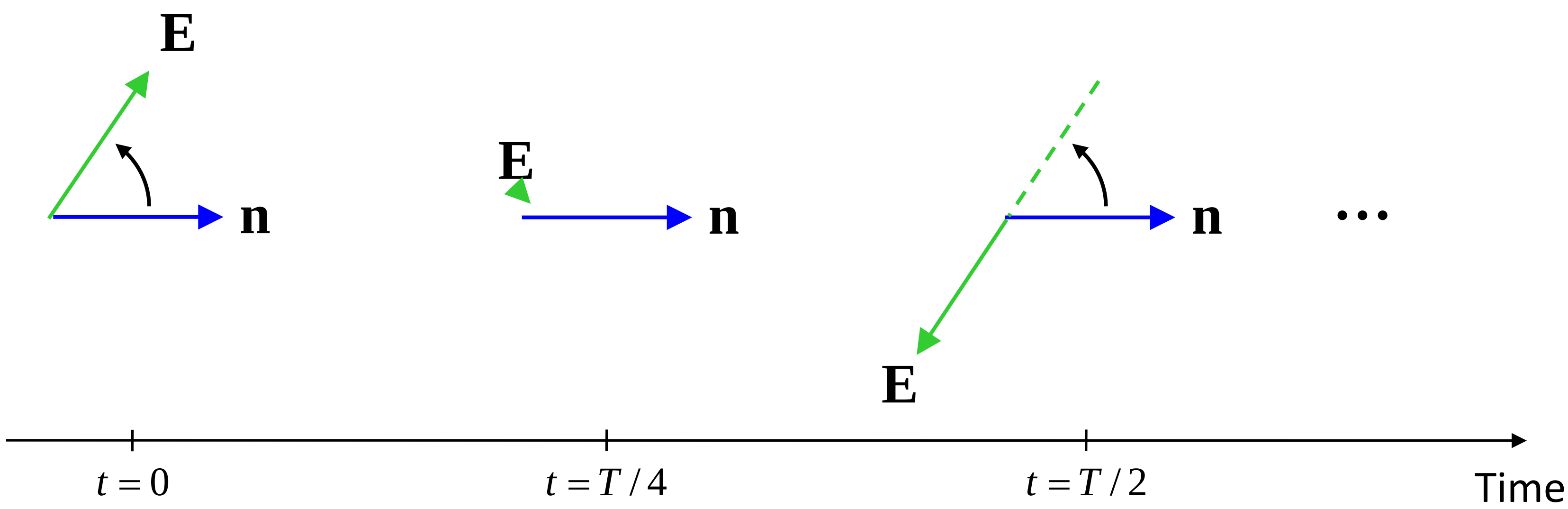
Two cases



$$\boldsymbol{\Gamma} = \varepsilon_0 \varepsilon_a (\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E})$$

Optical fields are time varying : so does the optical torque density

$$\mathbf{E}(z,t) = \mathbf{E}_0 e^{i(kz-\omega t)}$$



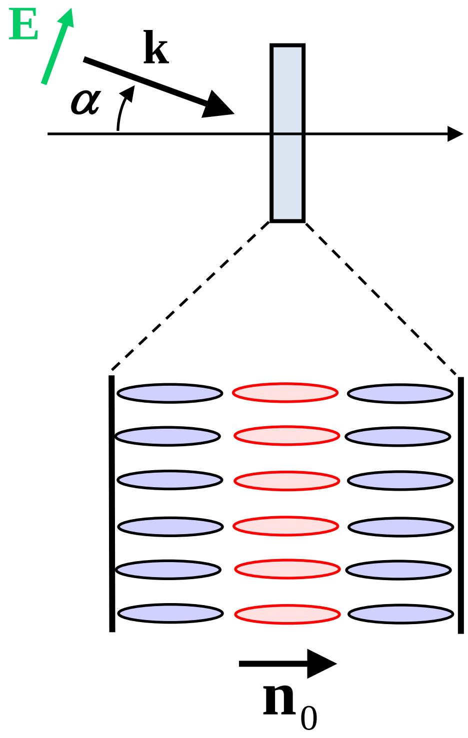
$$\boxed{\boldsymbol{\Gamma} = \varepsilon_0 \varepsilon_a \left\langle (\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E}) \right\rangle_t \neq 0}$$

Quadratic optical reorientation

Outline

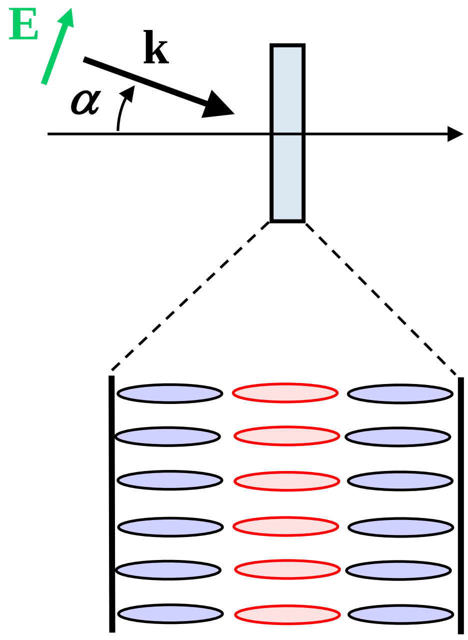
1. A (very) short introduction to liquid crystals
2. Dielectric and optical torques
3. **Orientational optical nonlinearities**
4. Role of the polarization state of light
5. Light-induced nonlinear rotations

A basic example of optical reorientation



$$\boldsymbol{\Gamma} = \varepsilon_0 \varepsilon_a (\mathbf{n}_0 \cdot \mathbf{E})(\mathbf{n}_0 \times \mathbf{E}) \neq 0$$

A basic example of optical reorientation

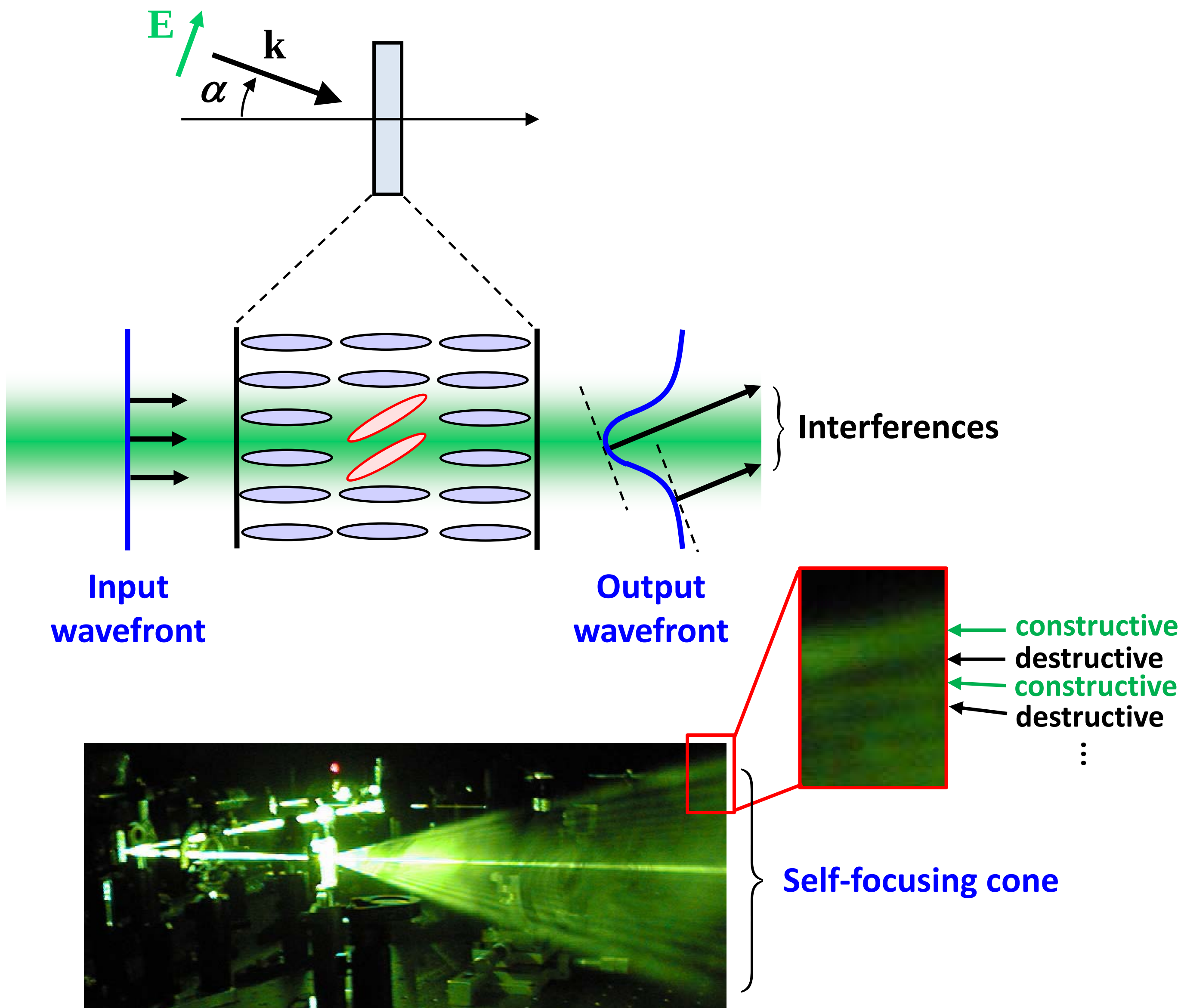


Collective reorientation

Compromise between optical and elastic torques

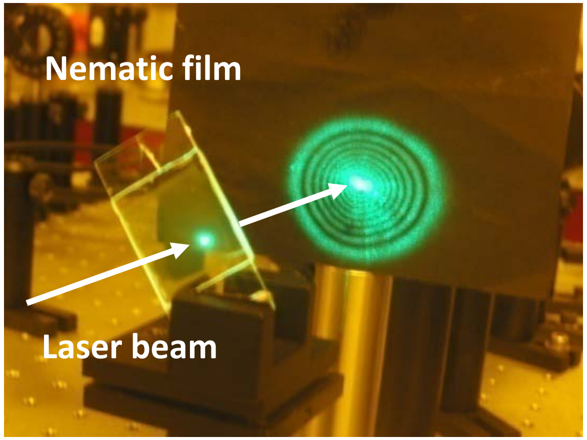
Which practical consequences ?

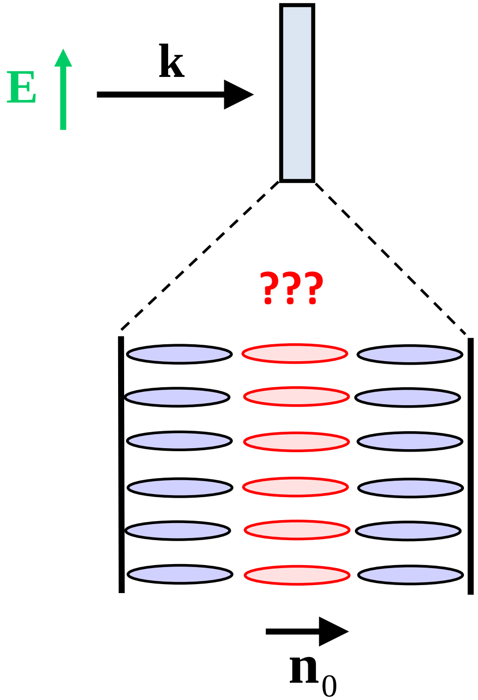
Giant self-phase modulation



A particular case : normal incidence angle

YES

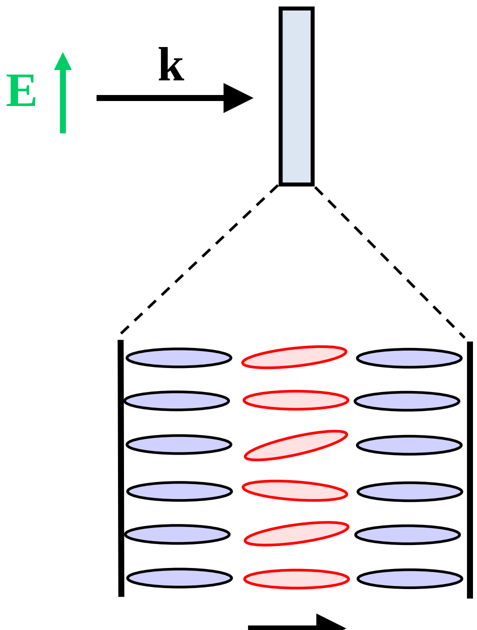




$$\mathbf{\Gamma} = \varepsilon_0 \varepsilon_a (\mathbf{n}_0 \cdot \mathbf{E})(\mathbf{n}_0 \times \mathbf{E}) = 0$$

Optical reorientation ???

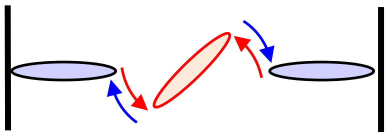
The role of thermal fluctuations



$$\mathbf{n}_0 + \delta\mathbf{n}$$

$$\boldsymbol{\Gamma} \approx \varepsilon_0 \varepsilon_a (\delta\mathbf{n} \cdot \mathbf{E})(\mathbf{n}_0 \times \mathbf{E}) \neq 0$$

Optical reorienting torque



Elastic restoring torque

Competition

$\Downarrow$

Orientational instability

Optical Fréedericksz transition

A. S. Zolot’ko *et al.*, JETP Lett. **32**, 158 (1980)

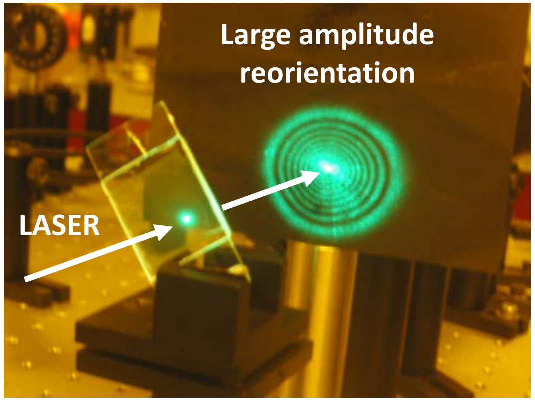
Outline

1. A (very) short introduction to liquid crystals
2. Dielectric and optical torques
3. Orientational optical nonlinearities
4. **Role of the polarization state of light**
5. Light-induced nonlinear rotations

Role of the polarization state of light : observations

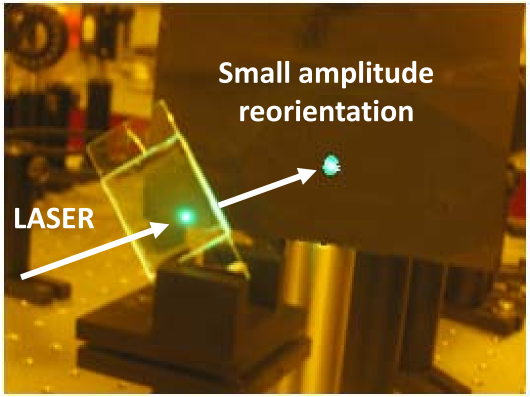
Just above the optical Fréedericksz transition

Linearly polarized light



A. S. Zolot’ko *et al.*, JETP Lett. **32**, 158 (1980)

Circularly polarized light

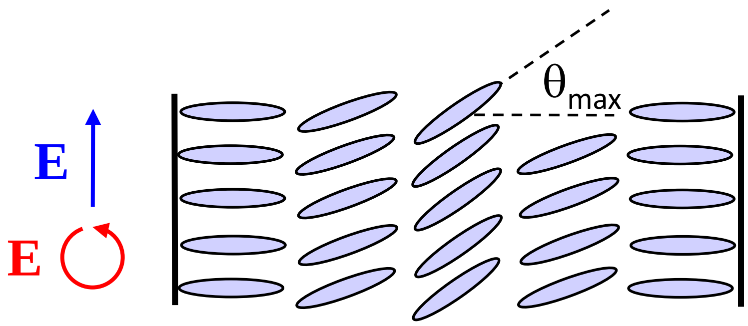


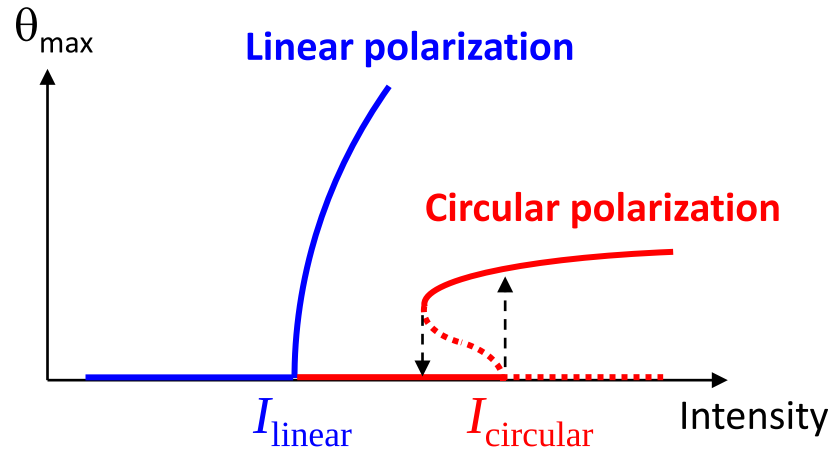
A. S. Zolot’ko *et al.*, JETP Lett. **34**, 250 (1981)

Trivial configuration

—

Non trivial behavior





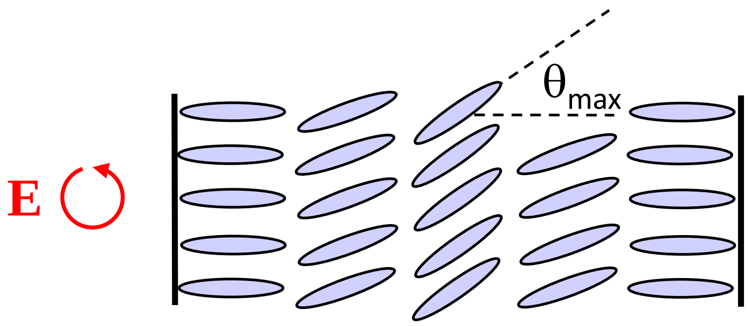
A few open questions

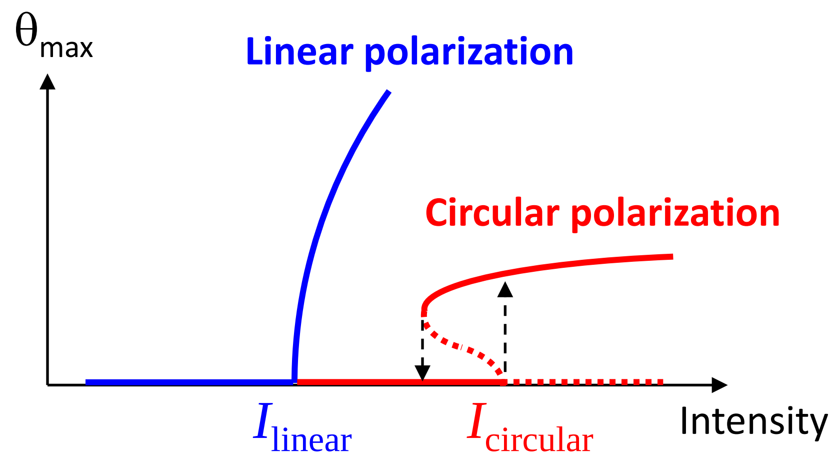
Q1. Twice higher threshold

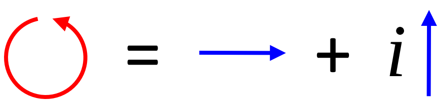
Q2. Optical phase locking

Q3. Existence of hysteresis

Role of the polarization state of light : reorientation diagrams







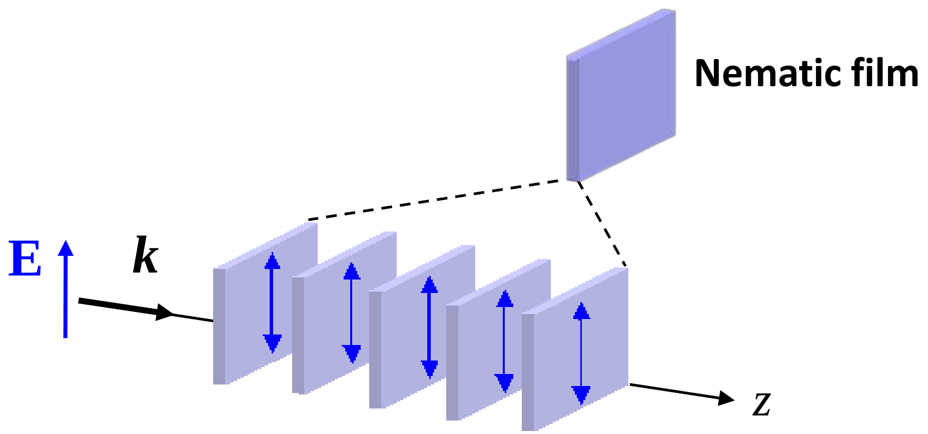
$$\Gamma = \varepsilon_0 \varepsilon_a (\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E})$$

Half the photons are not useful

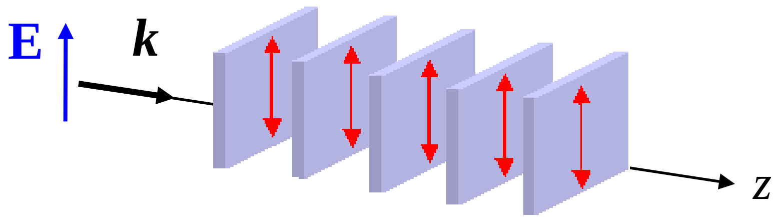
⇓

Need for twice more

A closer look to the propagation of light : linear polarization

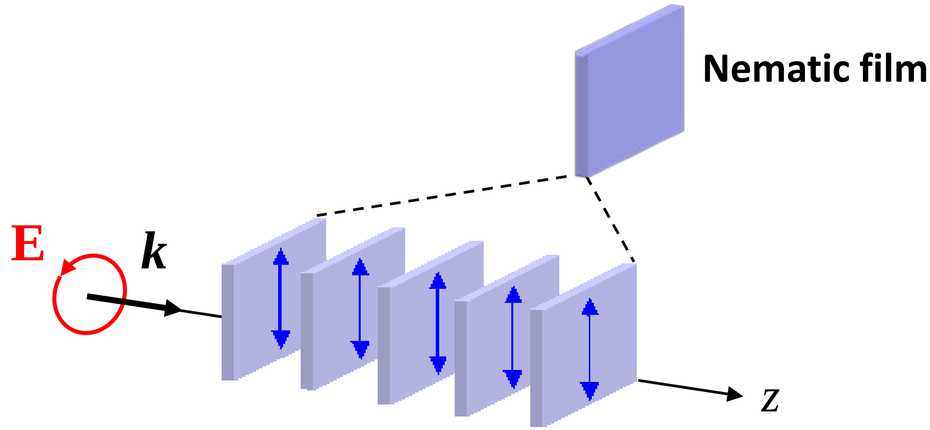


Neutral axes distribution : planar reorientation

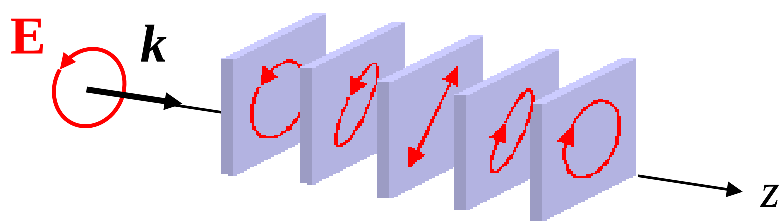


Polarization state distribution : input polarization state is preserved

A closer look to the propagation of light : circular polarization



Neutral axes distribution : ~~planar reorientation ?~~

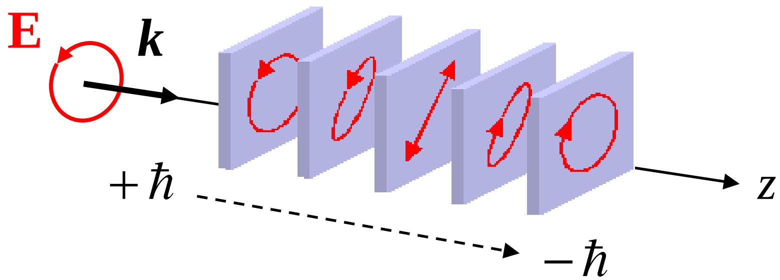


Polarization state distribution : spatial modulation !

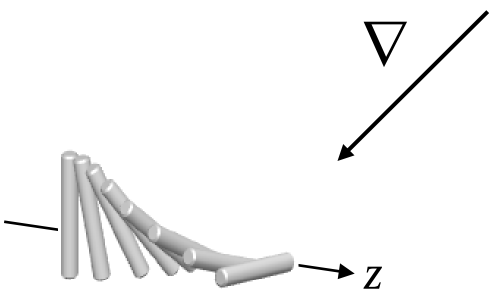
3D reorientation

A closer look to the propagation of light : circular polarization

Spin angular momentum of light : $s_z = \pm\hbar$

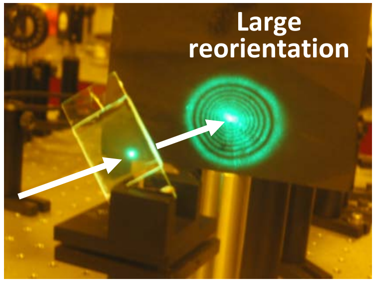


Spin angular momentum changes

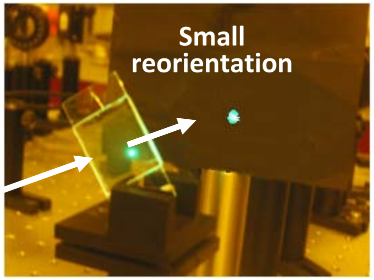


Torsional deformations

Non adiabatic propagation $\Rightarrow$ e/o coupling



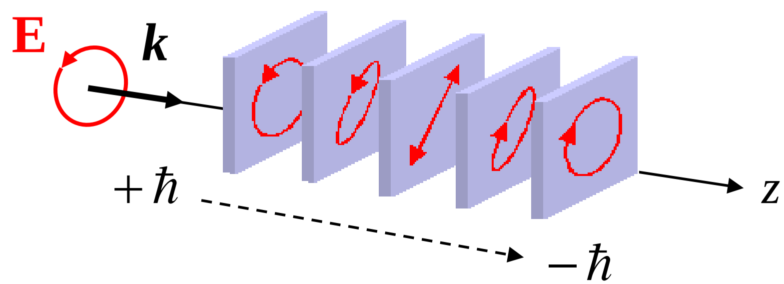
Linear input polarization



Circular input polarization

A closer look to the propagation of light : circular polarization

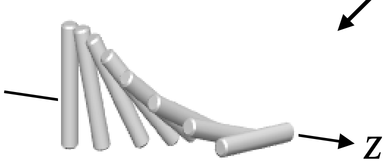
Spin angular momentum of light : $s_z = \pm\hbar$



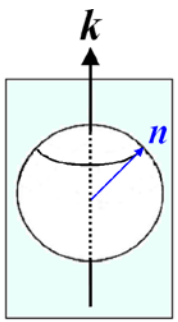
Spin angular momentum changes

∇

$\int$



Torsional deformations



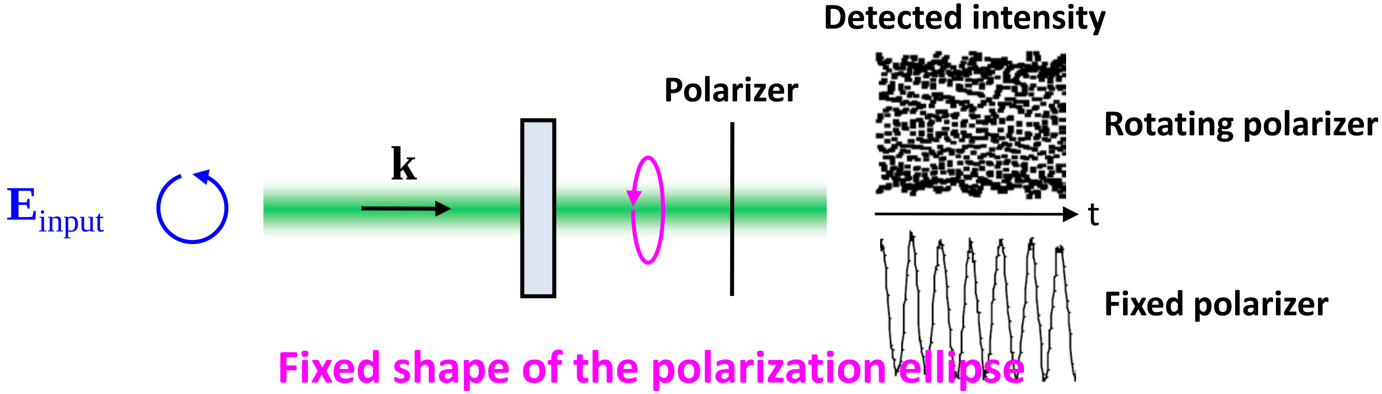
Collective molecular precession

Global rotation in the lab : molecular precession

Observed in 1986

A. S. Zolot'ko *et al.*, Preprint No 326, Lebedev Physics Institute, USSR Academy of Sciences (1986)

E. Santamato *et al.*, PRL **57**, 2423 (1986)



Fixed shape of the polarization ellipse

Time-dependent orientation of the polarization ellipse

Where is the fuel ?

Director precession

$$\omega \rightarrow \omega' < \omega$$

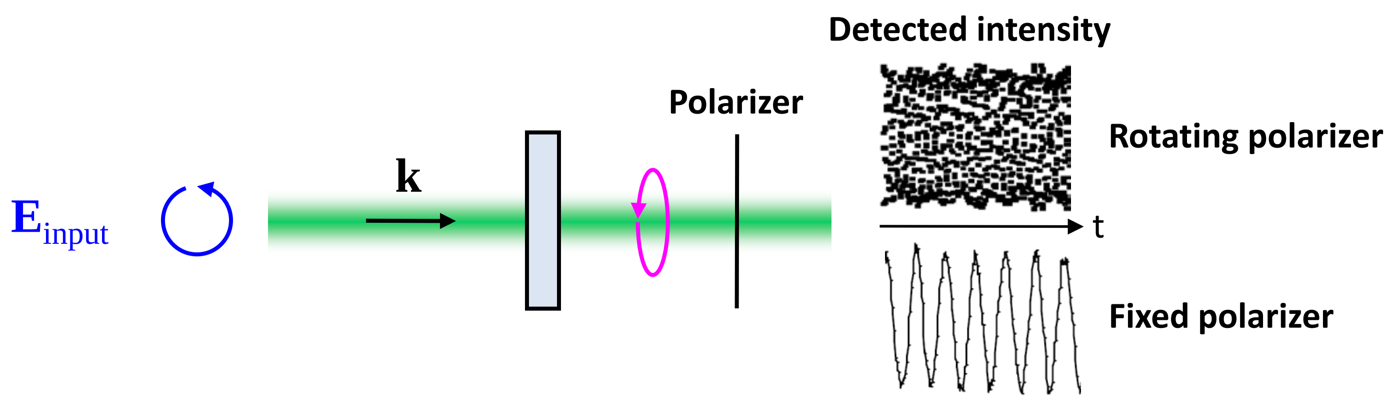
Redshift of the frequency of light

$$\delta\omega/\omega \sim 10^{-16} - 10^{-17}$$

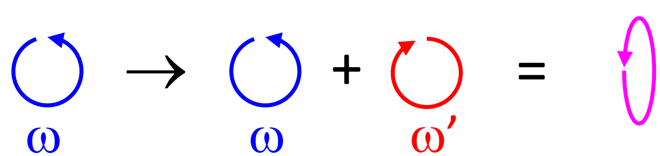
Observed in 1986

A. S. Zolot’ko *et al.*, Preprint No 326, Lebedev Physics Institute, USSR Academy of Sciences (1986)

E. Santamato *et al.*, PRL **57**, 2423 (1986)



Where is the fuel ?



$\Omega = (\omega - \omega')/2$

Polarization ellipse rotation frequency

Total spin angular momentum of light deposited per unit time

$$\Phi\,(s_3^{\text{in}} - s_3^{\text{out}})\,\hbar$$

Number of photons passing through per unit time

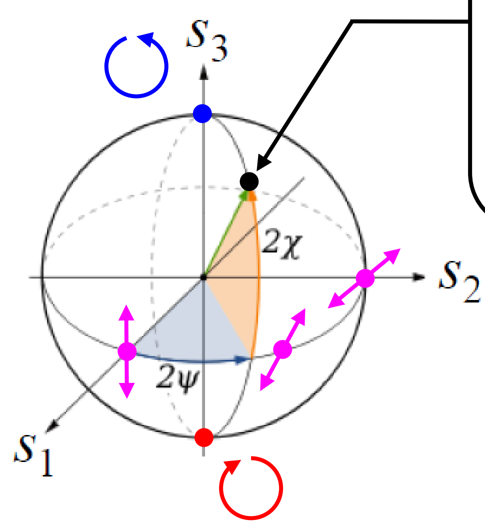
Reduced third Stokes parameter

Stokes parameters $\begin{cases} S_0 = I_x + I_y \\ S_1 = I_x - I_y \\ S_2 = I_{+45^\circ} - I_{-45^\circ} \\ S_3 = I_R - I_L \end{cases}$

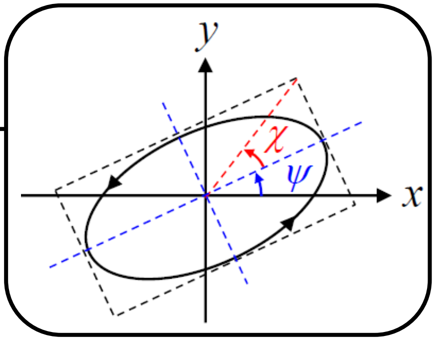
Reduced Stokes parameters $s_i = S_i / S_0$

$-1 \le s_i \le +1$

Poincaré sphere



Polarization state



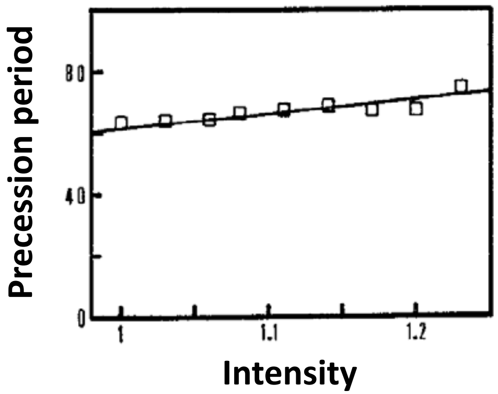
Total spin angular momentum of light deposited per unit time

$$\Phi\,(s_3^{\text{in}} - s_3^{\text{out}})\,\hbar$$

Number of photons passing through per unit time

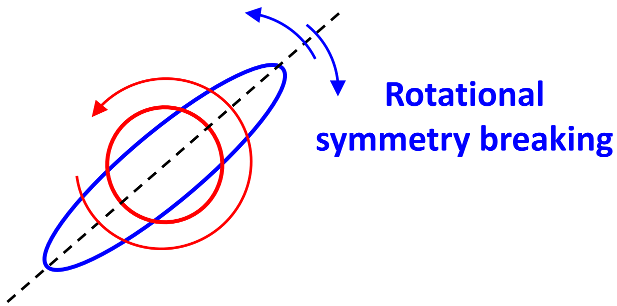
Reduced third Stokes parameter

Varying incident photon flux



L. Marrucci *et al*., PRA **46**, 4859 (1992)

Varying incident polarization state



E. Santamato *et al*., PRL **64**, 1377 (1990)

Control limited to ≈ 10%

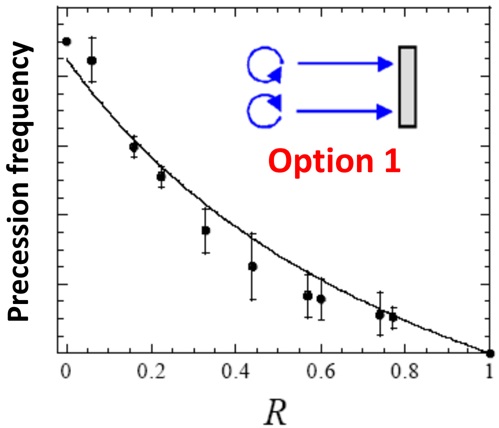
Controlling collective molecular precession

Total spin angular momentum of light deposited per unit time

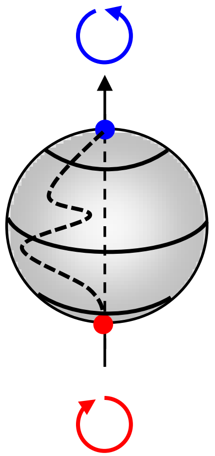
$$\Phi\,(s_3^{\text{in}} - s_3^{\text{out}})\,\hbar$$

Fixed photon flux + Rotational symmetry

T. V. Galstyan *et al.*, PRL **78**, 2760 (1997)



100% control "unstable"



---- Broken symmetry

—— Preserved symmetry

1. **Two co-propagating beams**
2. **Incoherent**
3. **Opposite circular polarization**
4. **Fixed total intensity**
5. **Varying intensity ratio R**

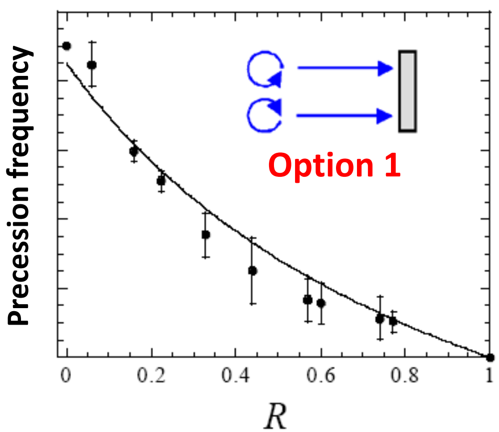
Controlling collective molecular precession

Total spin angular momentum of light deposited per unit time

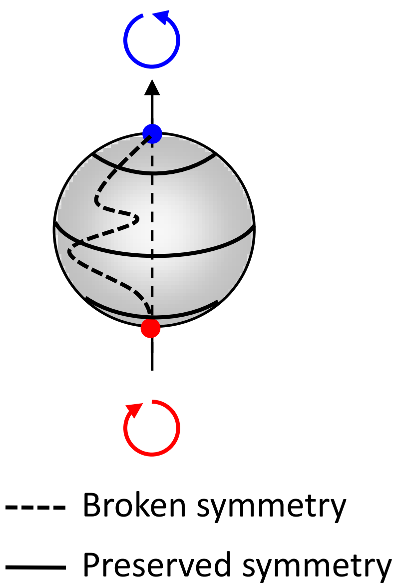
$$\Phi\,(s_3^{\text{in}} - s_3^{\text{out}})\,\hbar$$

Fixed photon flux + Rotational symmetry

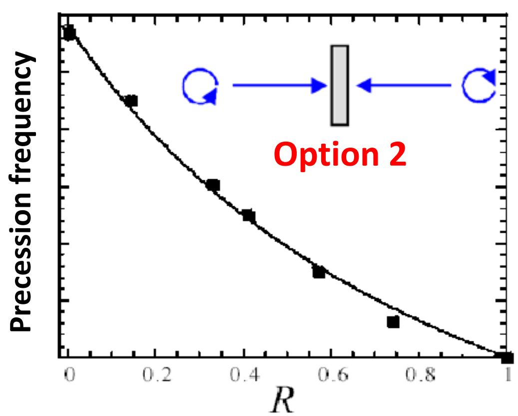
T. V. Galstyan *et al.*, PRL **78**, 2760 (1997)



100% control "unstable"



E. Brasselet *et al.*, Opt. Commun. **186**, 291 (2000)



100% control "stable"

Differential rotation in the lab : twisted distortions

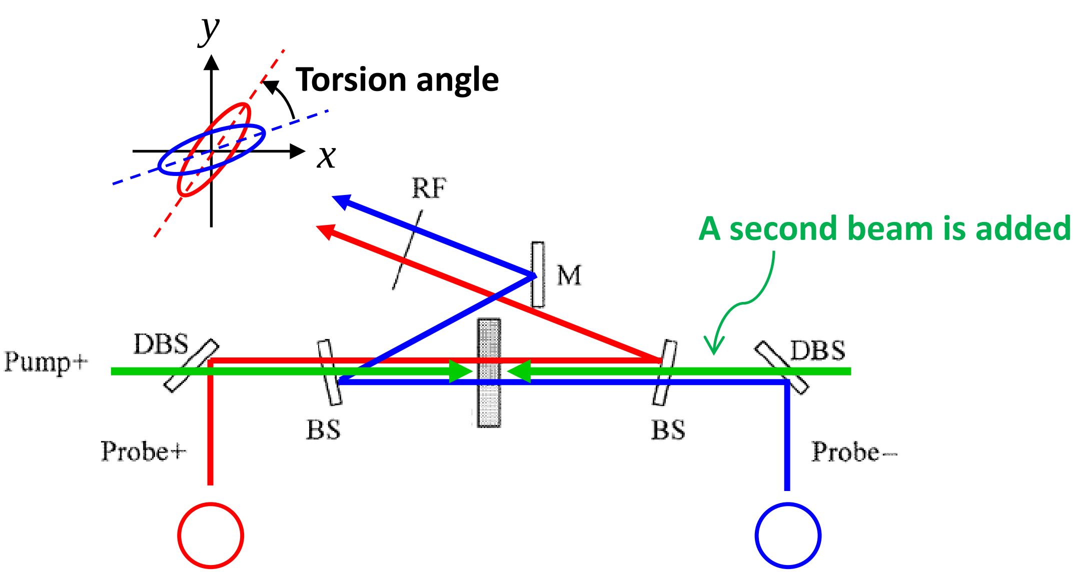
Predicted in 1990

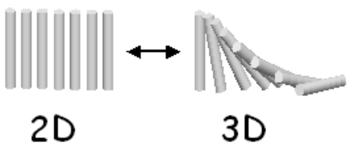
A. S. Zolot'ko *et al.*, JETP Lett. **34**, 250 (1990)

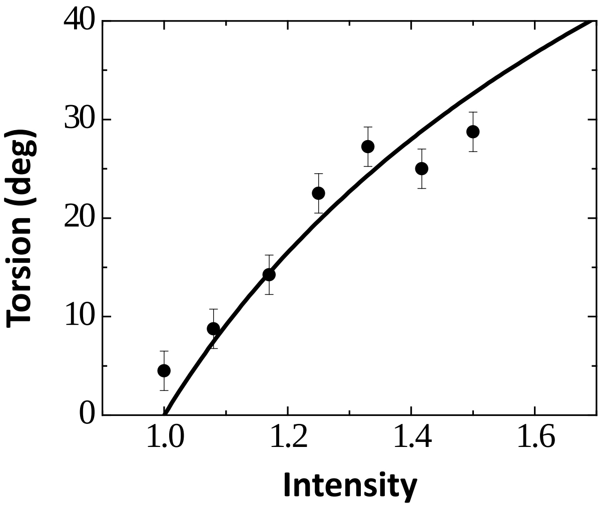
Observed and controlled in 2001

E. Brasselet and T. V. Galstian, Opt. Commun. **200**, 241 (2001)

Light-controlled chirality







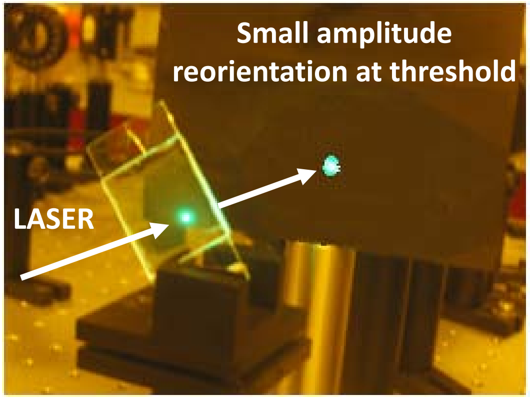
Differential polarimetry technique

Outline

1. A (very) short introduction to liquid crystals
2. Dielectric and optical torques
3. Orientational optical nonlinearities
4. Role of the polarization state of light
5. **Light-induced nonlinear rotations**

Nonlinear rotations under circularly polarized light

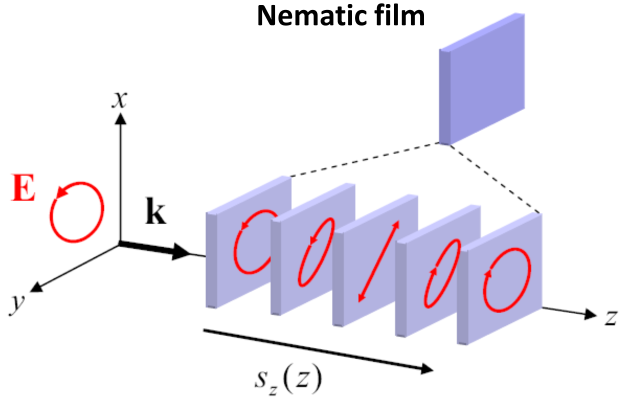
1980 Experimental discovery



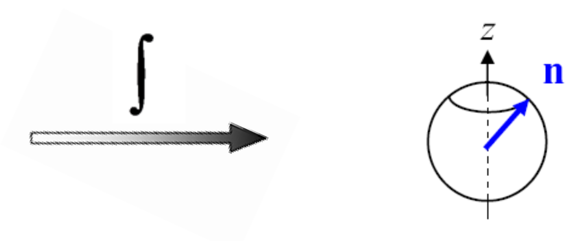
A. S. Zolot'ko *et al.*, JETP Lett. **34**, 250 (1981)

1980 **Experimental discovery**

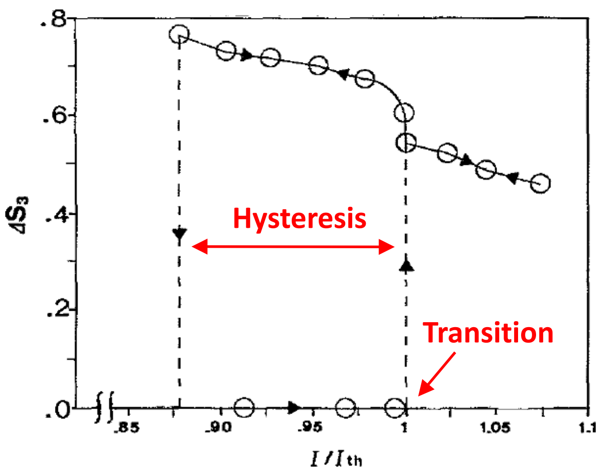
1986 **Collective molecular precession : experimental demonstration**



GLOBAL spin angular momentum transfer



Spin angular momentum transfer



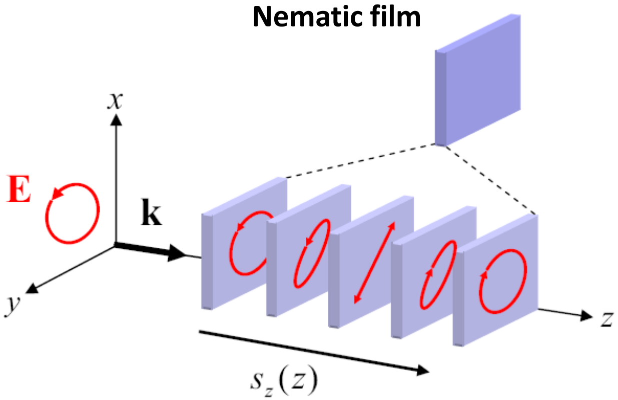
E. Santamato *et al.*, PRL **57**, 2423 (1986)

Nonlinear rotations under circularly polarized light

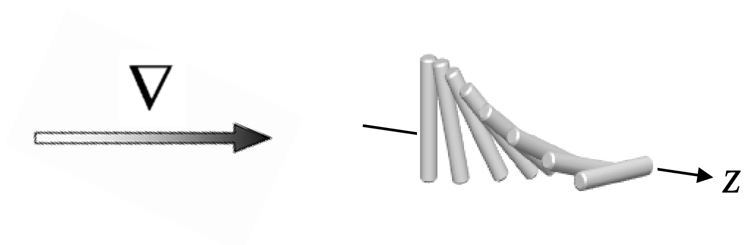
1980 **Experimental discovery**

1986 **Collective molecular precession : experimental demonstration**

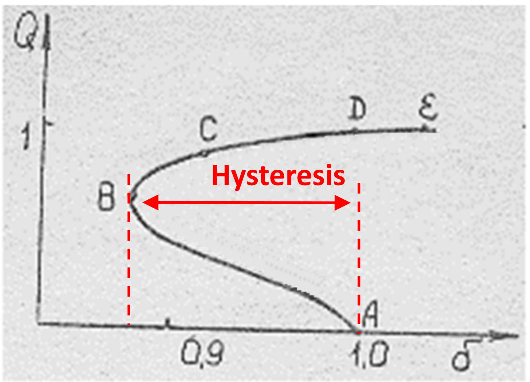
1990 **Near-threshold theoretical description : the role of twisted elastic distortions**



LOCAL spin angular momentum transfer



Reorientation amplitude vs. intensity



A. S. Zolot'ko *et al.*, JETP Letters **52**, 62 (1990)

1980 **Experimental discovery**

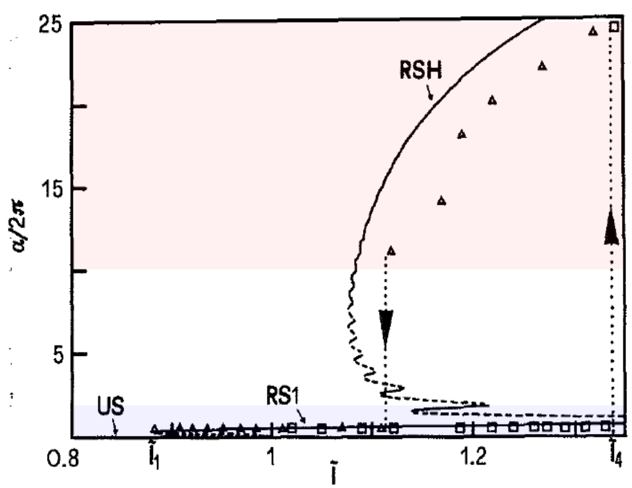
1986 **Collective molecular precession : experimental demonstration**

1990 **Near-threshold theoretical description : the role of twisted elastic distortions**

1992 **Behavior far above threshold**

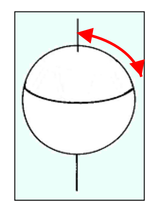
2001 **Observation of twisted elastic distortions**

Reorientation amplitude vs. intensity

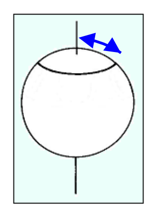


L. Marrucci *et al.*, PRA **46**, 4859 (1992)

?



"High amplitude" precession



"Small amplitude" precession

Nonlinear rotations under circularly polarized light

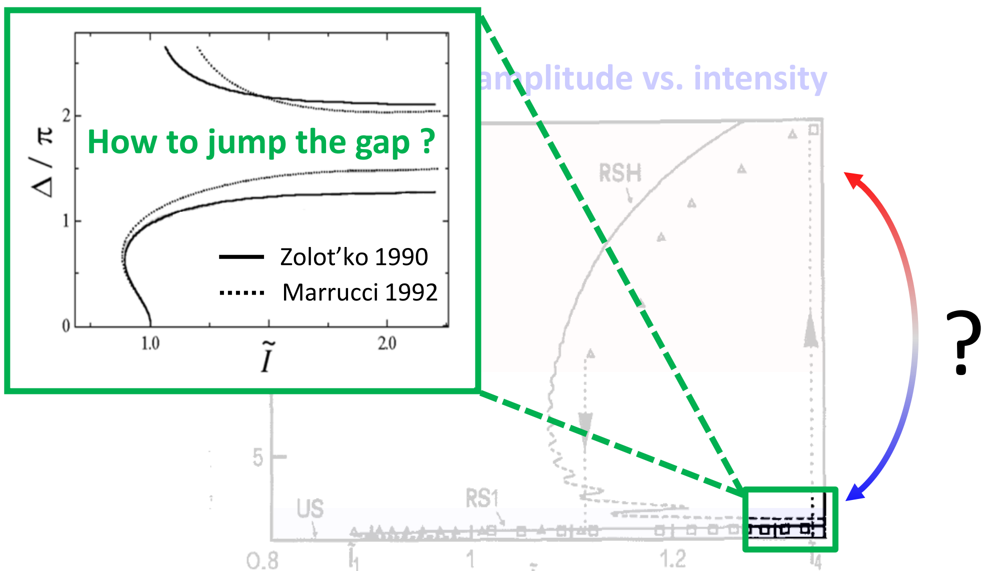
1980 **Experimental discovery**

1986 **Collective molecular precession : experimental demonstration**

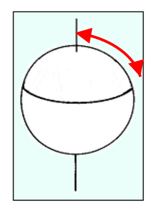
1990 **Near-threshold theoretical description : the role of twisted elastic distortions**

1992 **Behavior far above threshold**

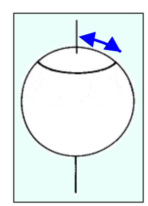
2001 **Observation of twisted elastic distortions**



L. Marrucci *et al.*, PRA **46**, 4859 (1992)



"High amplitude" precession



"Small amplitude" precession

Nonlinear rotations under circularly polarized light

1980 Experimental discovery

1986 Collective molecular precession : experimental demonstration

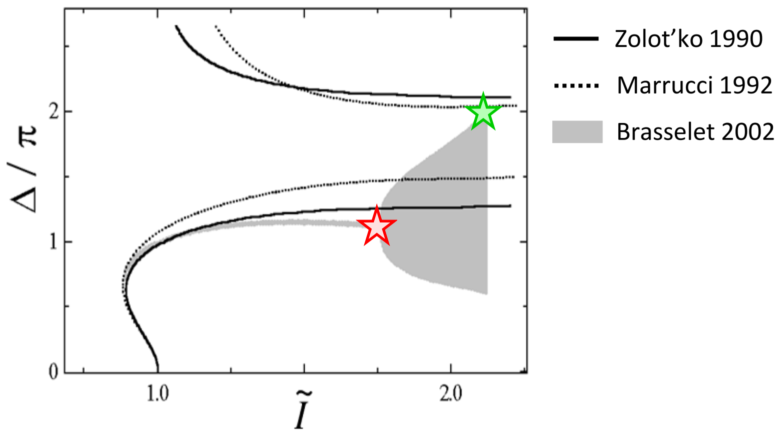
1990 Near-threshold theoretical description : the role of twisted elastic distortions

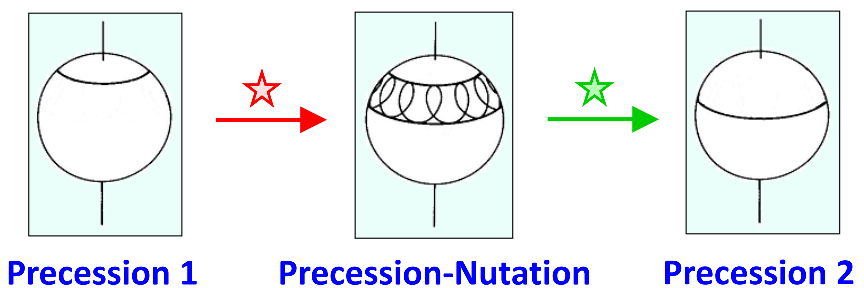
1992 Behavior far above threshold

2001 Observation of twisted elastic distortions

2002 Secondary instabilities : prediction and observation of molecular nutation

Reorientation amplitude vs. intensity

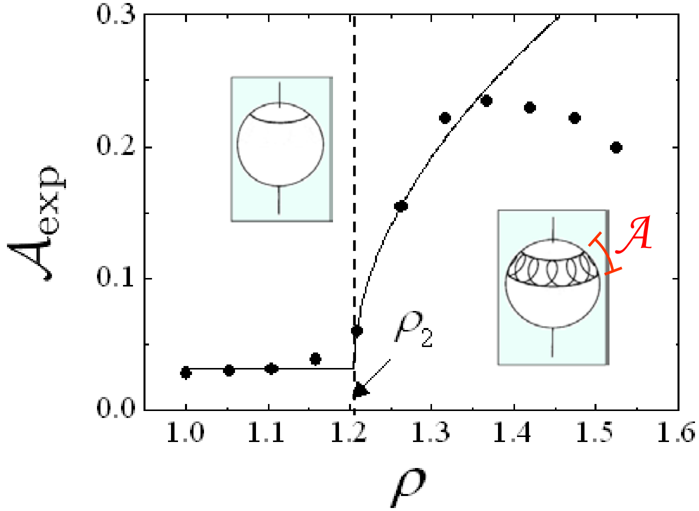




E. Brasselet *et al.*, Phys. Lett. A **299**, 212 (2002) ; PRE **67**, 031706 (2003)

Nonlinear rotations under circularly polarized light

Hopf bifurcation



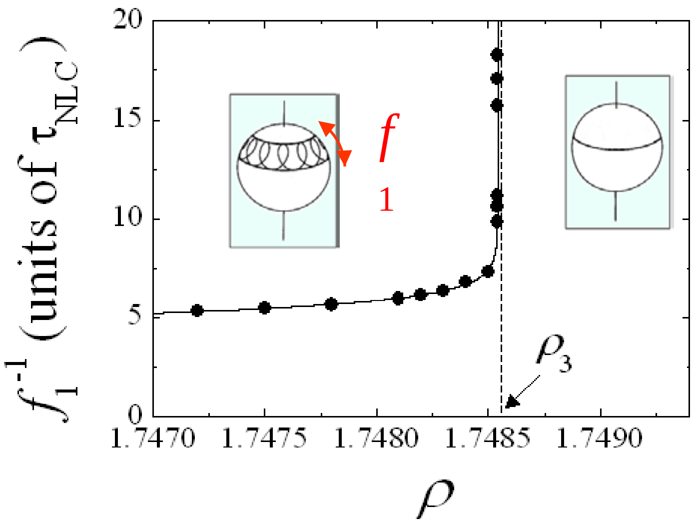
$\gamma_{\mathrm{exp}} = 0.46 \pm 0.08$

$\gamma_{\mathrm{theo}} = 1/2$

Nutation limit cycle amplitude

$$\mathcal{A}(\rho) - \mathcal{A}(\rho_2) = \mathcal{O}(\rho - \rho_2)^{\gamma}$$

Homoclinic bifurcation



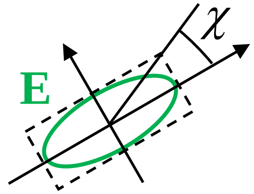
● calculated values

— best fit

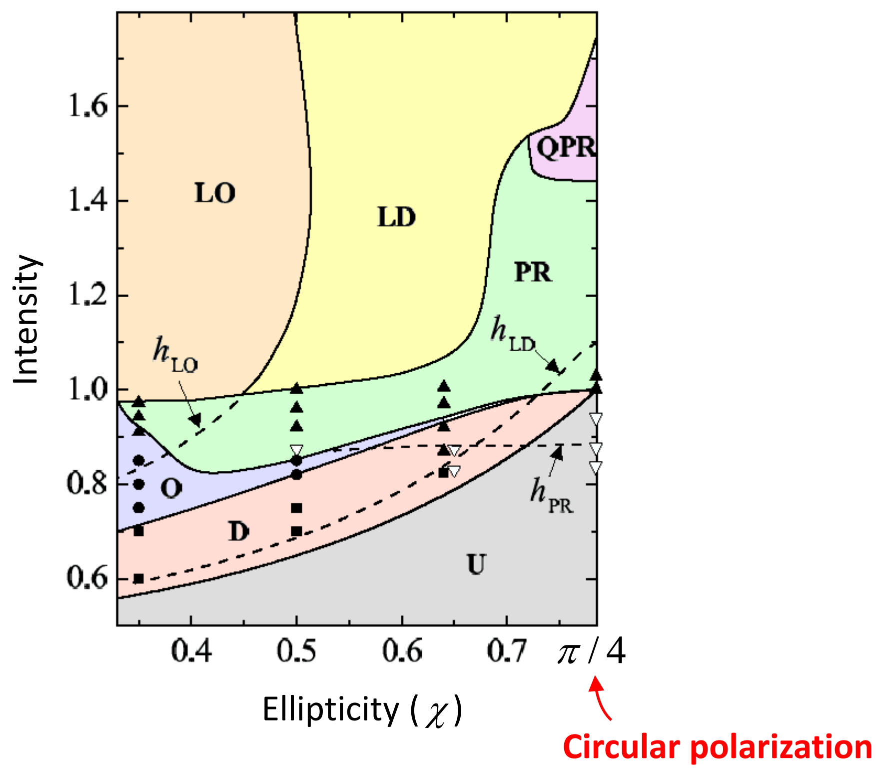
Nutation period divergence

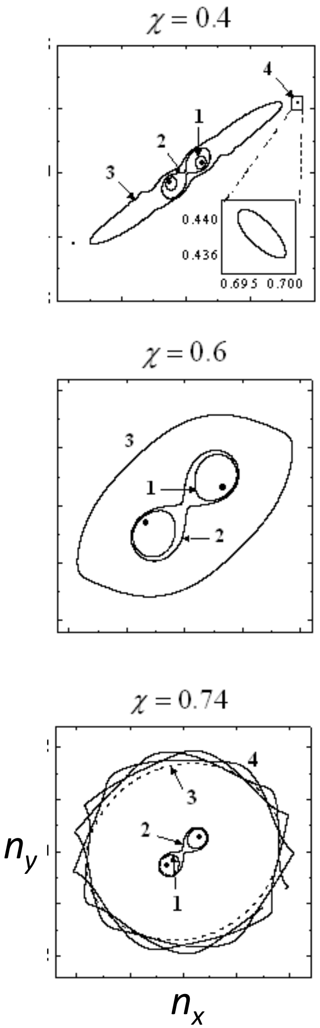
$$1/f_1 \propto \mathcal{O}\left[\ln(\rho_3 - \rho)\right]$$

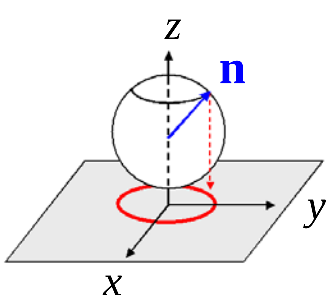
2005 **Complete theoretical description in the plane wave limit**



Map of dynamical regimes



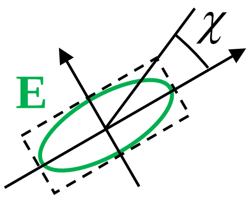




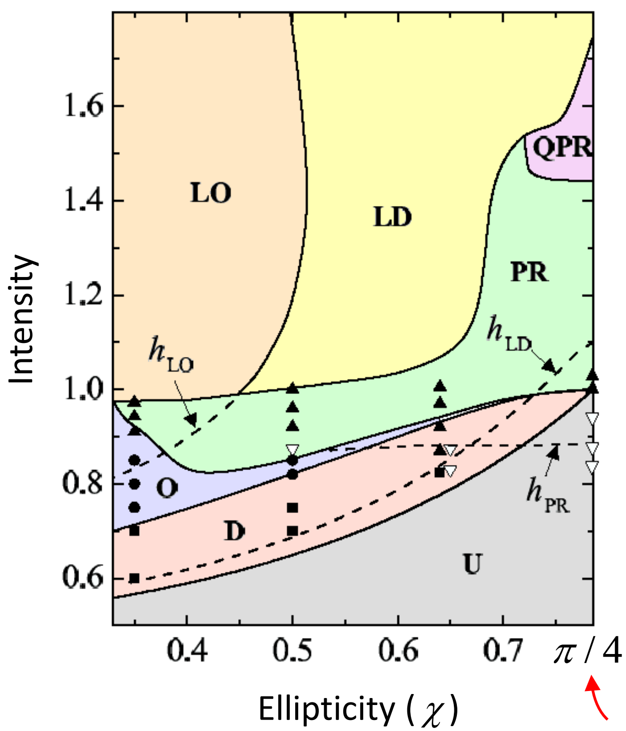
E. Brasselet *et al.*, JOSA B **22**, 1671 (2005)

D. O. Krimer *et al.*, JOSA B **22**, 1681 (2005)

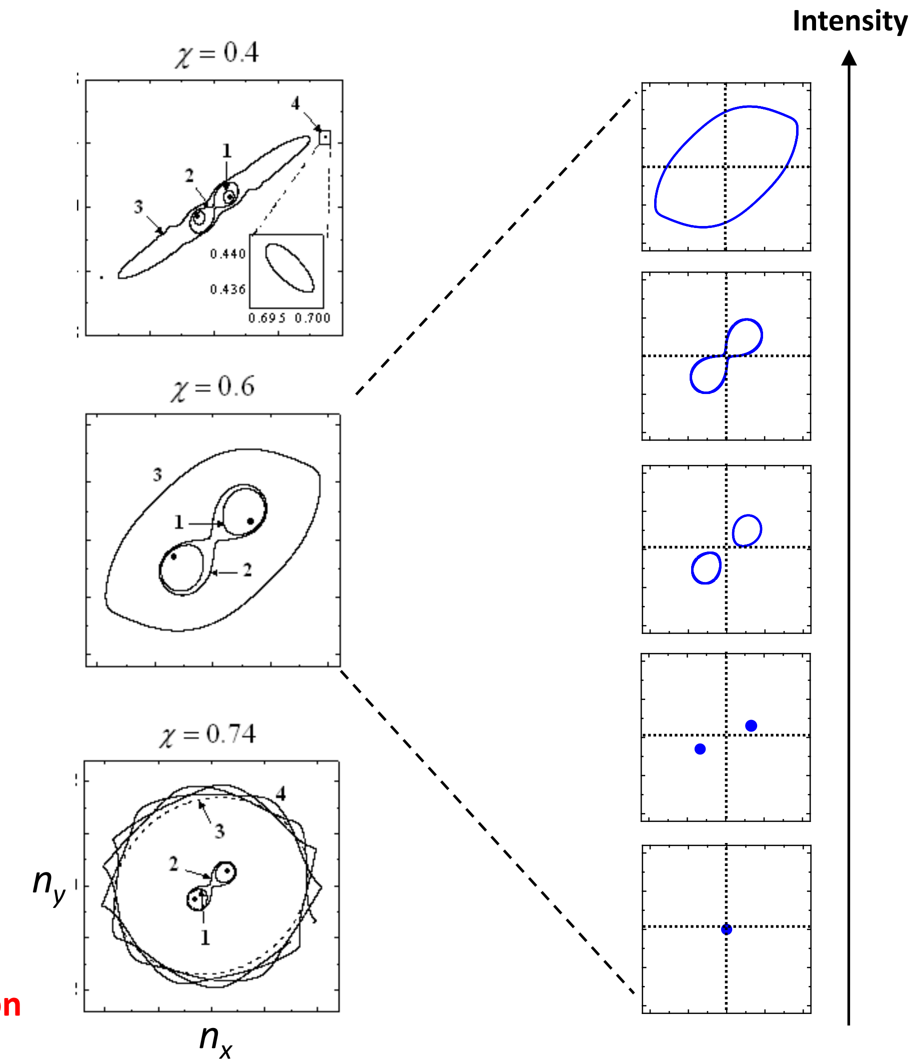
2005 **Complete theoretical description in the plane wave limit**



Map of dynamical regimes



Circular polarization

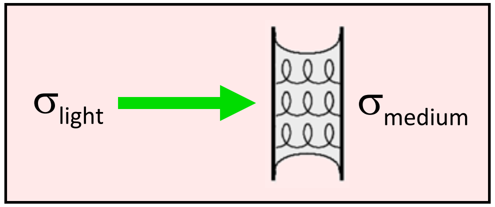


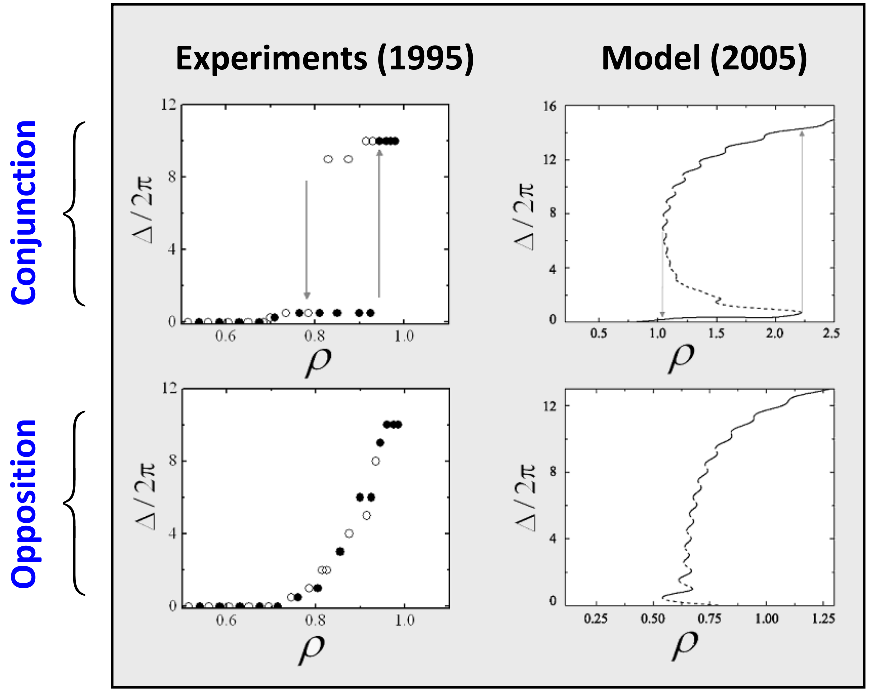
E. Brasselet *et al.*, JOSA B **22**, 1671 (2005)

D. O. Krimer *et al.*, JOSA B **22**, 1681 (2005)

2005 **Complete theoretical description in the plane wave limit**

Nematic doped by a chiral agent (cholesteric)





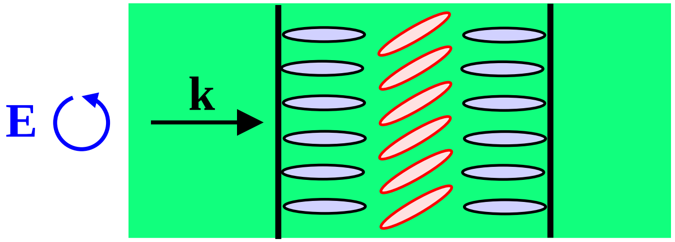
E. Brasselet *et al.*, Euro. Phys. J. E **17**, 403 (2005)

Translation symmetry breaking : emergence of complex dynamics

2005 **Complete theoretical description in the plane wave limit**

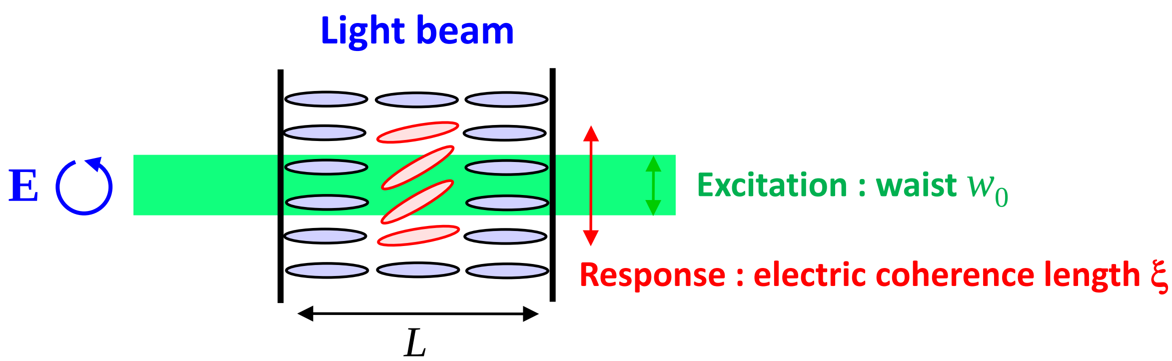
2006 **Finite beam size effects and nonlinear dynamics**

Plane wave



2005 Complete theoretical description in the plane wave limit

2006 Finite beam size effects and nonlinear dynamics



Criterion $\xi / w_0 \begin{cases} << 1 : \text{spatial locality} \\ \approx 1 : \text{spatial nonlocality} \end{cases}$

$$\frac{\xi}{w_0} = \frac{2}{\pi}\left(\frac{\varepsilon_{//}}{\varepsilon_{\perp}}\right)^{1/2} \frac{1}{\delta + \delta_c} \Rightarrow \begin{cases} \delta >> \delta_c : \text{locality} \\ \delta << \delta_c : \text{nonlocality} \end{cases}$$

$$\delta = 2w_0 / L$$

$$\delta_c = 2\sqrt{2} / \pi \approx 0.90$$

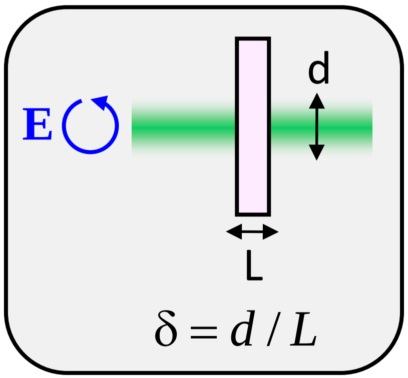
New dynamical regimes are expected

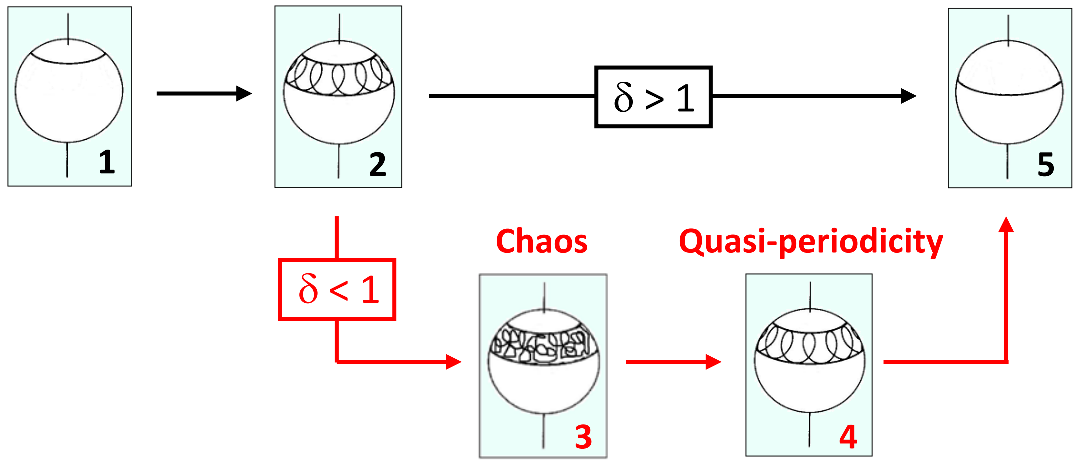
E. Brasselet *et al.*, JOSA B **23**, 36 (2006)

Translation symmetry breaking : emergence of complex dynamics

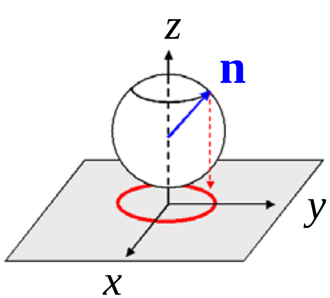
2005 Complete theoretical description in the plane wave limit

2006 Finite beam size effects and nonlinear dynamics

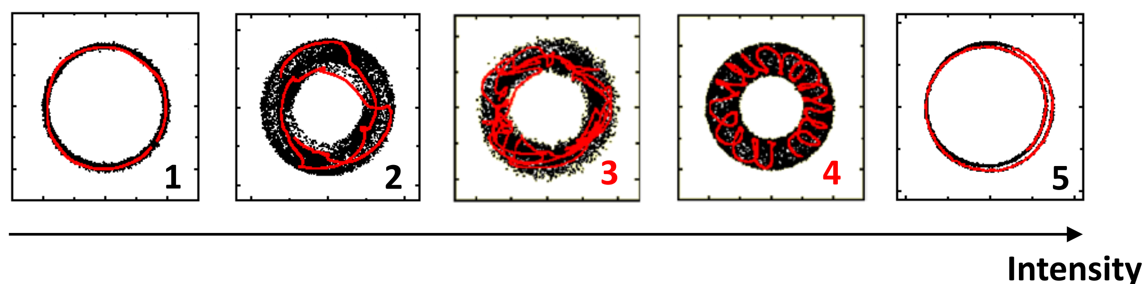




E. Brasselet *et al.*, Phys. Rev. E **73**, 021704 (2006)



Experimental scenario



Translation symmetry breaking : emergence of complex dynamics

2005 Complete theoretical description in the plane wave limit

2006 Finite beam size effects and nonlinear dynamics

2008 First 3D model that describes light-induced reorientation dynamics

E. Brasselet *et al.*, Phys. Rev. E **78**, 031703 (2008)

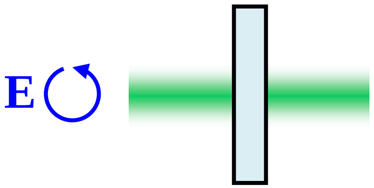
Optical reorientation under circular polarization

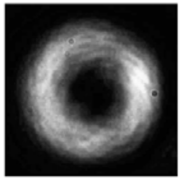
2005 **Complete theoretical description in the plane wave limit**

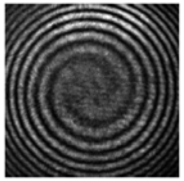
2006 **Finite beam size effects and nonlinear dynamics**

2008 **First 3D model that describes light-induced reorientation dynamics**

2009 **A novel phenomenon : topological optical reorientation (Lecture 4)**







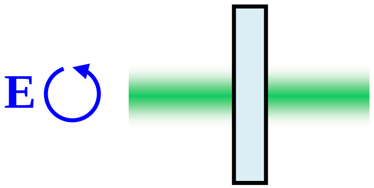
Optical reorientation under circular polarization

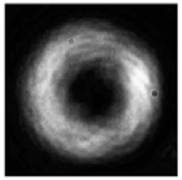
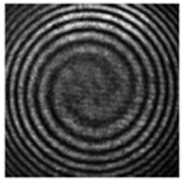
2005 Complete theoretical description in the plane wave limit

2006 Finite beam size effects and nonlinear dynamics

2008 First 3D model that describes light-induced reorientation dynamics

2009 A novel phenomenon : topological optical reorientation **(Lecture 4)**



Conclusion

Simple material systems but extremely rich light-matter interaction in presence of optical angular momentum